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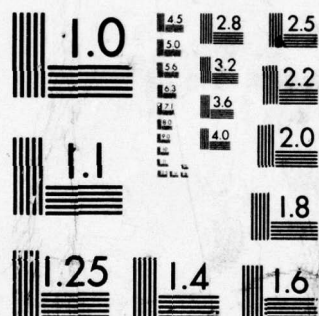
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PROJECT SQUID

SEMI-ANNUAL PROGRESS REPORT ✓

1 OCTOBER 1976

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PROJECT SQUID HEADQUARTERS ✓
CHAFFEE HALL
PURDUE UNIVERSITY
WEST LAFAYETTE, INDIANA

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SEMI-ANNUAL PROGRESS REPORT

PROJECT SQUID

A COOPERATIVE PROGRAM OF FUNDAMENTAL RESEARCH
RELATED TO JET PROPULSION
OFFICE OF NAVAL RESEARCH, DEPARTMENT OF THE NAVY

THIS REPORT COVERS THE WORK ACCOMPLISHED
DURING THE PERIOD 1 APRIL 1976 TO 30 SEPTEMBER 1976
BY PRIME AND SUBCONTRACTORS UNDER
CONTRACT N00014-75-C-1143, NR-098-038

1 OCTOBER 1976

PROJECT SQUID HEADQUARTERS
CHAFFEE HALL
PURDUE UNIVERSITY
WEST LAFAYETTE, INDIANA

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I. AERODYNAMICS AND TURBO

THREE DIMENSIONAL TRANSONIC FLOWS IN COMPRESSORS AND CHANNELS

The University of Michigan, Ann Arbor, Michigan
Subcontract No. 8960-10

Professor T.C. Adamson, Jr., Principal Investigator
Professor M. Sichel, Principal Investigator

Introduction

The calculation of mixed transonic flow fields in channels and rotors is a difficult task, either analytically or numerically. The complications arise from the fact that the mixed nature of the flow means that two dimensional approximations and those approximations introduced by linearization, are not valid in general. Furthermore, any shock waves which occur, do not fill the whole channel, but there can be considerable effects due to shock reflection in the part of the flow that is supersonic. The goals of the present study are first to ascertain those conditions under which solutions derived from linear governing equations are adequate to describe the flow in a rotor and to provide alternate methods of solution in those cases where they are not, and second, to provide approximate solutions for the flow structure in inlets and rotors which will be helpful in setting up three dimensional numerical solutions. The solutions are being obtained in the form of asymptotic expansions, with the first phase

of the work being the study of a flow problem which models the flow through a compressor rotor.

Discussion

The model problem chosen for study may be interpreted as the flow into a linear cascade with the blades aligned parallel to the incoming flow. The symmetry boundaries emanating upstream and downstream of each blade in such a case are replaced with walls, so that the flow field studied is the flow through a three dimensional channel with a constriction, this constriction corresponding to half blades on opposite walls. The variation in the rotor tangential velocity component with radius is accounted for by consideration of a linear gradient in the velocity entering the channel, while the thermodynamic properties in this incoming flow are held constant. The solutions found for this problem illustrate a very important physical concept: even though the flow through the nozzle-like constriction is mixed, choked flow can exist when the average Mach number at the minimum area is unity. This can occur only when the incoming velocity is restricted to a flow with a gradient in velocity, a condition which exists for the velocity relative to the blades in a rotor. The existence of such solutions was reported at the Workshop on Transonic Flow Problems in Turbomachinery held at the Naval Postgraduate School, Monterey, California, February 11-13, 1976.⁽¹⁾ The detailed development of these solutions, including the description of inner regions described in the following, will be submitted

for publication soon.

The solutions mentioned above have been derived from linear governing equations. However, there are various inner regions where nonlinear equations may be necessary, and it is these regions which have been studied during the past six months. The first of these regions exists about the leading edge of the blades, where, as long as noncusped leading edges are considered, the asymptotic expansions break down. In the subsonic region, this breakdown occurs because there is a stagnation region, where the flow velocity obviously cannot be described by a perturbation from the external flow velocity. In this case, there is apparently a logarithmic singularity at the leading edge, similar to that found in airfoil theory. In the supersonic part of the leading edge flow, there is a bow shock which reflects from the shock from the adjacent blade (at the centerline) and then reflects from the blade back toward the centerline, back to the blade, etc. That is, there must be a number of inner regions, each containing a shock wave. This means that the pressure distribution along the blade (constriction) surface is really discontinuous, jumping each time a wave strikes and reflects from the surface. However, in the asymptotic limit as the flow approaches sonic velocity and the shocks become weaker, and the reflections more numerous, the pressure distribution approaches the continuous distribution given in the solutions. It should be mentioned that this same problem and a similar result is found in numerical computations,⁽²⁾ where the bow shocks and reflected shocks are "smeared" out so that a continuous pressure distribution results.

There is another region where the solutions derived from linearized equations break down, and this is the region containing the plane where the cross sectional flow area is minimum, corresponding to the throat in a nonshear flow. From calculations made so far, it appears that it is not necessary to consider special inner solutions for this region if the outer solutions are interpreted properly.

Work is continuing on gaining an understanding of the solutions in the above mentioned inner regions because these solutions will be of the greatest help in setting up numerical computations and judging the limits of validity of analytical solutions.

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- (2) Oliver, D. and Sparis, P., "Computational Studies of Three Dimensional Transonic Shear Flow: Work in Progress," G.T.L. Report No. 101, Massachusetts Institute of Technology, 1970.

THE UNSTEADY RESPONSE OF AN AXIAL FLOW ROTOR
TO DISTORTED INFLOWS

Applied Research Laboratory
The Pennsylvania State University
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Subcontract No. 8960-4

Edgar P. Bruce, Principal Investigator

Introduction

The objective of this program is to analyze the time-dependent interaction between an isolated axial flow rotor and a spatially fixed circumferentially varying flow field. The major variables are reduced frequency, and rotor blade space-to-chord ratio, stagger angle, mean angle of attack, and design loading level. An outline of the experimental phase of the program is presented in Table 1. The experimental results are being used to provide design information and to assess the validity of theoretical models.

The experiments are being conducted in the ARL Axial Flow Research Fan (Reference 1). This facility has a hub radius of 12.06 cm (4.75 inches), a hub-to-tip radius ratio of 0.442, and operates in the subsonic incompressible flow regime. The rotor blades have a 10 percent thick C1 profile with a chord of 15.24 cm (6.00 inches) and a span of 14.99 cm (5.90 inches). One of the rotor blades contains a 2.54 cm (1.00 inch) span strain gauged segment centered on the mean radius. The time-dependent signals from the strain gauges are recorded on FM tape and are subsequently analyzed by a digitizing, phase-lock averaging process to determine

the segment unsteady normal force and pitching moment. As shown in Table 1, the first two phases of this program were devoted to an evaluation of rotor response, while the scope of the third phase has been expanded to include an evaluation of the effect of the rotor on the distorted flow field.

Discussion

The initial phase of this program has been completed (Reference 2). The experimental results have been compared with predicted results from two unsteady cascade theories -- the Whitehead-Smith theory and the Henderson-Daneshyar theory (References 3 and 4, respectively). The comparison shows that the Whitehead-Smith theory predicts the trend of the variation of all of the measured and computed quantities well and that the Henderson-Daneshyar theory predicts many of the trends shown by the data in regions where it is not influenced by divergent behavior near its resonance points.

Recent progress on this program consists of: 1) completion of the second phase testing and the related theoretical computations using the Henderson-Daneshyar and Whitehead-Smith theories, 2) initiation of an effort to extend the Henderson-Daneshyar theory to include prediction of the unsteady pitching moment, and 3) fabrication of the blades for the free-vortex loading distribution rotor required for the third phase testing.

The second phase test program was completed on August 2, 1976. These data are now being digitized and phase-lock averaged as required for data analysis. The portion of this program which consisted of a check of data taken during the initial test phase was expanded by including all three rotor configurations ($N = 4, 6, \text{ and } 12$) instead of

Table 1. Summary of the Scope of the Experimental Phase of the First Three Years of the ARL Project SQUID Research Program.

Time Period	Type of Rotor Blade	Parameters*	Objective	Status
10/1/73 to 9/30/74, Phase 1	Uncambered (with instrumented segment bounded by rectangular sections)	D=1,2,4 and 6 N=4,6 and 12 $\lambda=35,45$ and 55 deg $\bar{\alpha}=0,4$ and 8 deg	To measure rotor blade element unsteady force and moment	Completed (Reference 2)
10/1/74 to 9/30/75, Phase 2	Uncambered (with instrumented segment bounded by cylindrical sections)	D=9,12 and 15 N=4,6 and 12 $\lambda=35,45$ and 55 deg $\bar{\alpha}=0,4$ and 8 deg D=4 and 6** N=12 $\lambda=35,45$ and 55 deg $\bar{\alpha}=0,4$ and 8 deg	To measure rotor blade element unsteady force and moment	Testing completed in 2nd and 3rd quarters of 1976. Data reduc- tion now in progress
10/1/75 to 9/30/76, Phase 3	Uncambered and cambered (with instrumented segment bounded by cylindrical sections)	Uniform inflow N=9 $\lambda=45$ deg (uncambered) and 50.3 deg (cambered) $\bar{\alpha}=0$ and 8 deg D=1,2,4,5,6,9,12 and 15 N=9 $\lambda=45$ deg (uncambered) and 50.3 deg (cambered) $\bar{\alpha}=0$ and 8 deg	To measure rotor blade element unsteady force and moment and determine unsteady losses by analysis of time-mean data	Testing scheduled for 4th quarter 1976 and 1st quarter 1977

*D is the number of sinusoidal distortion cycles per circumference, N is the number of rotor blades,
 λ is the rotor blade stagger angle and $\bar{\alpha}$ is the rotor inflow mean angle of attack.

**This series of tests with the instrumented segment of the rotor blade bounded by cylindrical sections was
included for comparison of results with earlier results obtained by employing rectangular sections.

only $N = 12$ as shown in Table 1, and by extending the testing at $N = 12$, $\lambda = 55$ deg and $\bar{\alpha} = 0, 4$ and 8 deg to include tests with $D = 1, 2$ and 5 .

The effort in the near future will concentrate on reduction and analysis of the Phase 2 data, continued developemnt of the Henderson-Daneshyar pitching moment expression, and Phase 3 testing.

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2. Bruce, E. P. and Henderson, R. E., "Axial Flow Rotor Unsteady Response to Circumferential Inflow Distortions," Project Squid Technical Report PSU-13-P, September 1975.
3. Whitehead, D. S., "Force and Moment Coefficients for Vibrating Airfoils in Cascade," Aeronautical Research Council R & M 3254, February, 1960.
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INVESTIGATION OF THE EFFECTS OF HIGH AERODYNAMIC
LOADING ON A CASCADE OF OSCILLATING AIRFOILS

United Technologies Research Center
East Hartford, Connecticut 06108
Subcontract 8960-19

Franklin O. Carta, Principal Investigator
Arthur O. St. Hilaire

Introduction

The basic objective of this research program is to study the phenomenon of dynamic stall on a cascade of oscillating airfoils. Measurements are being made of the unsteady chordwise pressure distribution, and efforts are being made to detect the occurrence of boundary layer transition and separation on the surface of an oscillating cascaded airfoil operating near the static stall condition.

Program Review

Tests have been conducted in the linear, subsonic Oscillating Cascade Wind Tunnel (OCWT) to determine the effects of high aerodynamic

loading on cascaded airfoils. An initial test program was performed in which hot film responses under modest loading conditions were studied (Ref. 1). This document contains a detailed description of the facilities used. Improvements were made in the instrumentation system, as described in Ref. 2, and an extensive test program was completed at high loading, as described in Ref. 3. Although loading had some effect on the aerodynamic damping of the system, it was found that the interblade phase angle had the most significant effect on predicted stability.

Summary of Results to Date

Two reports have now been written to document the unsteady aerodynamics of a loaded oscillating cascaded airfoil in a subsonic stream. The first report (Ref. 1) was aimed at a preliminary study of the surface flow phenomena at low and moderate loading. The primary emphasis therein was an examination of the heat transfer response of the hot film transducers placed along the chord of the center airfoil of an eleven blade cascade, all of which were oscillated in pitch about their midchords. This first report also reviewed the history of the multi-blade stall flutter problem and it was noted that the state of the art in investigating the dynamic stall of cascaded airfoils lags significantly behind the comparable advances that have been made in isolated airfoil studies.

A second document (Ref. 3) reports on the work performed to date on the same cascade operated more deeply into the dynamic stall regime. The blades were oscillated about their midchords at frequencies up to 17 cps and at free stream velocities up to 200 ft/sec. The tests were conducted at interblade phase angles of 0 deg and 45 deg, and data were obtained for three values of mean incidence angle representing a range from modest to heavy aerodynamic loading. In addition to hot film measurements it was possible to obtain unsteady pressure distributions which were integrated to yield normal force, pitching moment, and work per cycle on the oscillating blade. It was observed that the interblade phase angle parameter had a significant effect on the stability characteristics of the cascade. Specifically, it was found that a change in interblade phase angle from 0 deg to 45 deg radically alters the character of the unsteady blade loading (which governs its motion in a free system) from stable to unstable. Furthermore, it was determined that the stability or instability is governed primarily by the phase angle of the pressure distribution over the forward 10 to 15 percent of the blade chord. Reduced frequency and mean incidence angle changes have a relatively minor effect on stability for the range of parameters tested.

Flow separation was detected using hot films and was visually observed during the test. The separation appears to be confined to a thin layer over the cascade blade suction surface. It occurs at high loading and exists over the entire chord at maximum oscillatory incidence angle.

Future Studies

Notification has been received from Project SQUID that this contract will be renewed this fall. In the next phase of this work we plan to include the following tasks in our program.

1. Perform additional tests at higher velocities and higher frequencies to extend the ranges of the relevant unsteady parameters.
2. Vary the interblade phase angle (simulating both forward and backward traveling waves) over a range of parameters to establish a cascade stability boundary dependent on this important variable.
3. Perform selected tests at increased gap/chord ratio to examine the effect of the proximity of neighboring blades on in-passage flow at high loading and during stall.
4. Introduce additional hot film instrumentation on the test blade and on blades adjacent to the test blade to examine a) spanwise flow effects, b) finite cascade effects, and c) blade-to-blade uniformity with respect to separation and reattachment trends of the boundary layer.
5. Perform additional flow visualization tests using tufts and high speed cinematography to examine the effects of these parameter and geometry changes on separation and reattachment.

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2. Project SQUID Semi-Annual Progress Report, 1 April 1976, pp. 8-19.
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AN INVESTIGATION OF PRESSURE FLUCTUATIONS AND STALLING
CHARACTERISTICS ON ROTATING AXIAL-FLOW COMPRESSOR BLADES

Virginia Polytechnic Institute
and State University, Blacksburg, Virginia
Subcontract No. 8960-13

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Introduction

This research program is principally involved with the study of unsteady phenomena in axial-flow compressors associated with stalling behavior. Special instrumentation for on-rotor pressure measurements has been developed and employed in a continuing experimental program. A companion effort to build an analytical flow model capable of including stalled flow, three-dimensional and unsteady effects has been pursued.

During this report period, pressures on rotor blades have been measured with a system designed for very accurate measurement of mean pressure levels. Comparisons with the flow model showed good agreement. A method for the inclusion of end-wall effects was developed and incorporated in the analysis. Predictions were checked with

measurements in a stationary cascade. A distorted inflow study was conducted using screens of varying circumferential extent. A new calibration system for on-rotor pressure transducers was designed.

Discussion

Mean Pressures on Rotating Blades

The on-rotor instrumentation developed previously under the program employs miniature, blade-mounted pressure transducers and a telemetry data transmission system. Although this system is capable of measuring and transmitting mean pressures, it was designed primarily for the fluctuating components.

A new on-rotor system, designed specifically for the measurement of mean pressure, has recently been installed on a low-speed (2400 rpm) research compressor. The new system employs a selecting valve (Scanivalve^R) with a single, stationary-mounted pressure transducer. Up to 36 rotor blade surface pressures are sensed through static taps arranged along the chord at three different span positions on the suction and pressure sides of the blades. An experimental investigation of the mean pressures on the surface of the rotor blades at various flow rates, including stall, has been completed with the new system. The experimental pressure distributions have been compared with the analysis developed under the program. The analysis

includes a combination of two-dimensional calculations connected through simple radial equilibrium. Displacement effects of the blade boundary layers are included in the potential flow calculation. The boundary layer calculations are based on a two-dimensional integral technique. For the low-speed compressor, the flow is assumed incompressible. Agreement between theory and analysis is quite good.

End-Wall Effects on Cascades

Discrepancies between measured pressure distributions on stationary cascades and those calculated by potential flow are assumed to be caused by boundary layers on the blades and the cascade end walls. An attempt to eliminate the effect of end-wall boundary layers is often made in cascade studies, but the effect is not completely eliminated and is known to be an important factor in compressor performance. Thus, an investigation of the effect of end-wall boundary layers has been conducted under the present program.

The chordwise pressure distribution was determined experimentally on a stationary cascade of blades that have the same cross section, stagger angle, aspect ratio (length/chord) and spacing (solidity) as the rotor blades in the low-speed compressor. The blade boundary layer was measured at the trailing edge and the end-wall boundary layer was measured upstream and downstream of the cascade. Several inlet flow angles corresponding to measured flow rates for the compressor were investigated.

A two-dimensional analysis, which has been developed under the program and includes the blade boundary layers, was modified to include the end-wall effects. The end-wall boundary layer is calculated with an integral method using the calculated pressure distribution. The boundary layer is assumed two-dimensional based on a pressure that is averaged between blades, but includes the effect of converging or diverging free stream lines. The pressure distribution is calculated from potential flow, accounting for the displacement effect of the boundary layers. Agreement between experiment and analysis was substantially improved by including the end-wall effects in the analysis.

Distorted Inflow Studies

The Scanivalve measurement system installed on the low-speed (2400 rpm) research compressor was used for a study of stalling behavior with circumferential inlet distortions. Although the system was essentially suited for mean or average pressure measurements, it was determined that response to pressure fluctuations extended to 75 Hz. This frequency response was adequate to study the onset of stall, and the reduction of stall margin by the screen-induced distortion.

A yaw-probe was used to determine the flow directions and velocities immediately in front of the single-stage rotor. To produce a more uniform distortion for study, screens were placed immediately ahead of the inlet guide vanes of the machine, producing a

relatively well-defined square-wave distortion pattern. Flow directions and velocities, and overall machine performance were determined for undistorted inlet flow, and with constant density screens of 45, 90, and 180 degree circumferential extent.

It was found in both undistorted and distorted inflow tests that the onset of stall could be detected by the appearance of a strong component at approximately $1/2$ rotor frequency in the analysis of the on-rotor pressure data. During normal operation, relatively small components at multiples of rotor frequency were apparent. As stall developed, the $1/2$ rotor frequency component appeared and remained as the machine was loaded to produce a deeper stall and large, wide-frequency band pressure fluctuations.

Using the frequency analyzer to determine the onset of stall, it was determined that the 45 degree screen had essentially no effect on the stalling point of the machine. The 90 and 180 degree screens had the effect of precipitating stall at lower rotor blade loadings. The inlet pressure profile, the downstream pressure, and the stall point information were used in a modified stall margin reduction formula, based on parallel compressor theory. A critical distortion extent (maximum reduction in stall margin) of about 60 degrees was predicted.

Study of the data from the experiment is continuing, with emphasis on the relationship of dynamic pressure profiles on the rotor blades to the critical distortion phenomenon. To effectively study the rapidly varying profiles, high response transducers and automatic

data reduction techniques will be required.

On-Rotor Calibration Techniques

Pressure transducers and the data transmission system on the compressor rotor must be calibrated and rechecked frequently to insure accurate results. In the past, the measurement system has been calibrated using water manometers and calibration resistors. Calibration has been done before and after each test run. A new system design permits the electrical calibration of each transducer at any time during a test run, without stopping the rotor. A calibration circuit is activated optically, without contact with the rotor, which sequentially generates a reference level and applies a calibration resistor to each of the transducers. The overall system gain can thus be checked at any time. A breadboard model of the calibration circuit has been successfully tested, and it is planned to package the components for mounting on compressor rotors.

Future Plans

Future efforts under the program will continue to be directed toward a better basic understanding of the unsteady effects associated with stall.

The analytical effort will be a fundamental approach using the present steady flow analysis as a base. Unsteady effects will be included in the boundary layer calculations and in the inviscid

flow analysis. As a first approach, it will be assumed that the flow is the same in all blade passages, which might be a reasonable approximation for surge. At a later time, the possibility for different flows in adjacent blade passages, which is necessary for description of inlet distortion and rotating stall, will be included.

The future experimental effort will include an investigation of the unsteady effects caused by a rapidly varying flow rates in the low-speed compressor. The flow rate will be varied by a large, quick-acting valve near the discharge, and the variations will be large enough to cause transient stall on the blades. The response of the flow over the blades will be studied with on-rotor measurements of the fluctuating pressures. Distorted inflow experiments will be continued, with emphasis on dynamic pressure variations in the distorted region.

EFFECTS OF TURBULENCE ON FLOW THROUGH AN
AXIAL COMPRESSOR BLADE CASCADE

Colorado State University
Fort Collins, Colorado

Subcontract No. 8960-15

Professor W. Z. Sadeh
Principal Investigator

This research program is concerned with the determination of the role played by outer turbulence in reducing the aerodynamic losses of a blade cascade at moderate Reynolds numbers. In particular, the management of turbulence for forestalling the occurrence of laminar separation and for yielding a profile turbulent boundary layer is the prime aim of this research endeavor.

During the current year only a very limited preliminary study has been undertaken since the principal investigator was on leave from Colorado State University till September 1976. The main goal of this effort has been to lay down the necessary background work for carrying out the experiments of phases I and II. In these two phases the effects of oncoming turbulence on the flow around a circular cylinder and about an isolated airfoil are to be investigated. The design of both bodies and of the three-dimensional array of hot-wire probes has been completed. A preliminary experiment will be conducted in the wind tunnel within the forthcoming three to four months.

An exploratory effort concerning the matching of the outer and inner solutions of the vorticity amplification theory has been initiated. This study concentrates on the evolution of turbulent energy at scales susceptible to undergo amplification as the turbulence is conveyed toward the body. Currently, several numerical schemes are being examined for ascertaining their capabilities for resolving this issue.

Semi-Annual Progress Report

FUNDAMENTAL RESEARCH ON RELAMINARIZATION PHENOMENA AS PRODUCED IN NOZZLES AND TURBINES

Southern Methodist University, Dallas, Texas
Subcontract No. 8960-6

Professor Roger L. Simpson, Principal Investigator
Associate Professor Michael A. Collins, Associate Investigator
Mr. C. R. Shackleton, Research Assistant

Introduction

Under a strong acceleration, a turbulent boundary layer can achieve a laminar-like character with the accompanying lower heat transfer rate of a laminar flow. Under such acceleration conditions in nozzles and turbines a reduced heat transfer is desirable. While several previous investigations have provided mean velocity profile and heat transfer information, little detailed flow structure information has been previously obtained. The objectives of the current research are to make measurements of the turbulence structure, especially of the wall "bursting" and wall spanwise vortex behavior, in relaminarizing flows. Results from this work

for two relaminarizing flows, each with a constant acceleration parameter $K (\equiv \nu U_\infty^{-2} dU_\infty/dx)$ have been reported in a Technical Report (1). Turbulence measurements were made using a single slanted hot-film probe and obtaining data at different probe orientations. The wall measurements were made using flush surface hot-film sensors. A more severely accelerating flow in which K increases linearly has been recently examined and research on a nozzle type K distribution is nearing completion.

The wall "bursting" and wall spanwise vortex behavior is believed to play an important role in the relaminarization process. In the viscous sublayer of a turbulent boundary layer, previous researchers have found that instabilities occur and that a "burst" of turbulence results for each instability. When relaminarization occurs, the bursting rate is expected to decrease to zero.

Discussion

In our last two Semi-Annual Reports, dated October 1, 1975, and April 1, 1976, the significant findings of our research up to that time were summarized. Considerable effort had been spent in developing the split-film hot-film technique for making measurements in very thin boundary layers.

Since our last report, we have used the split-film in making measurements in the strongly accelerated flow where K increased

linearly. A procedure for correcting the data for velocity gradient effects is being examined since this effect is extremely important near the wall. Similar measurements are being obtained for the nozzle type K distribution.

The governing equations for the model of Hanjalif and Launder (2) for low Reynolds number turbulent flow are being programmed for comparison with the results from this experimental program.

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II. COMBUSTION AND CHEMICAL KINETICS

A SHOCK TUBE STUDY OF H_2 AND CH_4 OXIDATION WITH N_2O AS OXIDANT

University of Missouri, Columbia, Missouri
Subcontract No. 8960-21

Anthony M. Dean, Principal Investigator
Don C. Steiner, Research Assistant

Introduction

The study of oxidation reactions in shock tubes has been stimulated by the advent of fast, accurate numerical integration routines. No longer tied to the steady state assumption, kineticists can more definitively test various oxidation mechanisms by a detailed comparison of calculated and observed concentration-time profiles. Unfortunately, application of this approach to even such "simple" systems as $CH_4/O_2/Ar$ is severely limited by the lack of reliable rate constant data at higher temperatures—the number of variables is simply too large. Recent work in this laboratory (1-3) has demonstrated that N_2O is a particularly useful precursor of oxygen atoms. These observations suggested that the study of oxidation reactions where N_2O replaced O_2 as oxidant might provide useful information about the rate of reactions of atomic oxygen with various molecules at high temperatures. The use of N_2O as an oxidant has several advantages; the most significant is that oxygen atom reactions will occur in an environment substantially free of O_2 . Successful completion of the N_2O studies would then permit one to approach the CH_4/O_2 system with prior knowledge of the rates of the oxygen atom reactions to be

encountered there. This reduction in the number of unknown variables should then allow for much more incisive testing of the oxidation mechanism.

Discussion

Our efforts to date have been focused upon four different areas of the program:

1. Development of a Method to Quantitatively Observe Oxygen Atoms. Our earlier work showed that shock heating of N_2O resulted in production of large concentrations of oxygen atoms with much smaller amounts of NO and O_2 produced in secondary reactions. Here the evidence was indirect—the time dependence of oxygen atom production was monitored and the effect of added H_2 upon the N_2O decay rate was observed. In the last few months we have obtained more direct evidence to document the utility of N_2O as a precursor of atomic oxygen. The "flame-band" emission technique used to monitor atomic oxygen was independently calibrated by shocking $H_2/O_2/CO/CO_2/Ar$ mixtures. This system rapidly achieves a state of "partial equilibrium" (4) wherein the concentrations of O and CO (responsible for the flame-band signal) are easily calculated using well known equilibrium constants. The calibrated detector was then used in further studies of the N_2O/CO system. The measured concentration of oxygen atoms in these experiments were in good agreement with that predicted in our earlier studies; only slight rate constant adjustments were required to quantitatively fit the new data. These developments were particularly encouraging since we now not only have added confidence in our source of oxygen atoms, but we can monitor the absolute oxygen atom con-

centration in future work. This will significantly simplify the kinetic analysis.

2. Development of the Procedures Required to Obtain Rate Constants in the $N_2O/H_2/CO$ System. The analysis of the observed flame-band profiles must take into account the effect of background emissions as well as the "averaging" produced by the finite slit width. The necessary correction procedures have been developed and tested to handle these experimental perturbations. These procedures are now routinely used in the data reduction process and should be directly applicable to most of our future work.

3. Experimental Observations on the $N_2O/H_2/CO/Ar$ and $O_2/H_2/CO/Ar$ Systems. Although our ultimate objective is a more detailed analysis of the oxidation of methane, we feel that it is prudent to direct our initial efforts at the oxidation of hydrogen. Unlike the case of methane, the H_2/O_2 system has been well characterized and the rate constants for individual reactions are reasonably well known. Thus we can use this system as a "calibration" device; analysis of the $N_2O/H_2/CO/Ar$ studies should yield values of $O + H_2 \rightarrow OH + H$ in good agreement with those attained from more traditional studies of the $O_2/H_2/Ar$ system. Agreement of our results with the literature here would suggest that our forthcoming work on the $N_2O/CH_4/CO/Ar$ system should yield equally reliable results for the reaction $O + CH_4 \rightarrow CH_3 + OH$. The studies of $O_2/H_2/CO/Ar$ were done to provide further information upon the reaction $OH + CO \rightarrow CO_2 + H$ which will be needed for our future work. At the same time knowledge of this reaction allows the rate constant for $O + H_2 \rightarrow OH + H$ to be determined more precisely. In these studies we have monitored both the oxygen atom and carbon dioxide concentration. Preliminary analysis of these observations suggests that the systems are behaving as expected.

4. Measurement of the Recombination Rate of Carbon Monoxide and Atomic Oxygen.

The recombination of carbon monoxide and atomic oxygen to form carbon dioxide was expected to be of some significance to our experiments because of the relatively large concentrations of these species. The literature data on this reaction is not consistent, and we thus initiated a series of experiments to directly measure the rate of this reaction. This rate is also of interest in the much larger context of dissociation-recombination theory. Our observations demonstrated that measurements of this reaction are remarkably sensitive to very small concentrations of impurities. Analysis of the data under conditions where such impurity effects were minimized yielded quite precise values of the recombination rate between 2100 and 3200 K. This value is consistent with a "normal" activation energy for CO_2 dissociation and suggests that many of the earlier measurements of CO_2 dissociation were affected by impurities.

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KINETICS OF PHASE CHANGE AND ENERGY EXCHANGE

Yale University, New Haven, Connecticut
Subcontract No. 4965-16

J. B. Fenn, Principal Investigator
M. Sinha and N. Lee

Introduction

The exchange of energy during collisions between molecules and the exchange of mass between condensed and gaseous phases often play important roles in energy conversion systems. We have been studying the kinetics of these processes by means of molecular beam techniques. In the past our primary concern has been with energy exchange in gas-gas molecular encounters. We are now devoting more attention to the rates and mechanisms of mass and energy exchange between phases. In particular, we are interested in what happens when molecules leave or arrive at liquid surfaces and in the clustering of molecules during the initial stages of nucleation-condensation.

The principal investigative tool in these studies is a free supersonic jet issuing from a small sonic nozzle into vacuum. Such jets provide extremely rapid cooling and are capable of achieving steady state conditions which are far removed from equilibrium. In addition, they are useful sources for molecular beams of high intensity in the interesting energy range below a few tens of electron volts.

Discussion

A. Precondensation Polymerization Kinetics in Gases. Having successfully calibrated the mass spectrometer for the determination of neutral dimer concentrations in freely expanding jets we studied dimer formation kinetics in argon, carbon dioxide and oxygen. We were able to fit the results rather well with a model based on a three body collision rate for molecules whose behaviour can be approximated by a Lennard-Jones 6-12 potential. We found an apparent activation energy in each case for dimer stabilization by a third monomer molecule corresponding to about 0.38 times the binding energy of the dimer, i.e. the depth of the attractive well. In addition we inferred the existence of a steric factor which has a value of 0.18 for argon, 0.22 for oxygen and 0.46 for carbon dioxide.

We were unable to extend our calibration procedure to the quantitative determination of trimer concentration. On the basis of scaling laws for relative terminal concentrations of trimer we were able to conclude that the formation of an argon trimer from a dimer also required a third body to remove the binding energy. In the case of carbon dioxide it appears that a trimer with fairly long term stability can be formed from a simple two body collision between a dimer and a monomer. We tentatively attribute these differences in behaviour between argon and carbon dioxide to the presence of more internal degrees of freedom in the latter. The binding energy has more ways to distribute itself among the various modes of motion in the molecule and the probability that it will concentrate in a single bond and rupture it becomes much less. Therefore, the lifetime of the complex is long enough so that it can be

cooled by subsequent two body collisions.

B. Phenomena at Liquid Surfaces. Experiments on the scattering of molecular beams from liquid surfaces were renewed after a period of suspension. We confirmed our earlier findings that the flux distribution of argon atoms scattered from a clean glycerine surface depends upon the incident energy. At 0.06 eV the distribution is diffuse with a pure cosine distribution. At 0.36 eV it becomes lobular with the peak at slightly less than the specular angle. At 0.44 eV the peak is at an angle slightly greater than the specular value. Similar results have been found with CO_2 though even at the lowest energy there does seem to be a small departure from a cosine distribution in the flux reflected at large angles.

We are now able to detect an enhancement of evaporative flux of substrate glycerine occasioned by the incident beam. As might be expected the enhancement increases as the energy of the incident molecules increases. Somewhat suprising to us is the fact that both argon and helium molecules from a room temperature source (at about 0.06 eV) also cause a small but definite increase in the glycerine signal from the mass spectrometer detector. In our earlier observations, which were much more crude, we had seen an apparent increase in evaporation when accelerated argon molecules struck the surface but not when they came from a room temperature source. These observations invite contemplation of what happens when molecules of equilibrium vapor strike a surface.

We think this enhancement of apparent evaporation rate may be due to a sputtering effect. We plan to study in some detail the spacial distribution of the evaporating flux and its dependence upon energy, mass and structure of the incident beam molecules. We also plan to look at liquids other than glycerine.

HIGH-TEMPERATURE FAST-FLOW REACTOR CHEMICAL KINETICS STUDIES

AeroChem Research Laboratories, Inc., Princeton, NJ 08540
Subcontract 8960-16

Arthur Fontijn, Principal Investigator
William Felder, Physical Chemist

Introduction

Reliable quantitative knowledge of the kinetics of free metal atom and metal oxide species is required for a good understanding and description of (i) the burning of metallized propellants and (ii) the exhaust properties of rockets using such propellants. Suitable techniques for obtaining such kinetic information were unavailable until we adapted the tubular fast-flow reactor technique to reach temperatures up to 2000 K (1). This development has made an essentially room temperature technique capable of being used for making measurements in the temperature range of conventional high-temperature techniques such as flames and shock tubes.

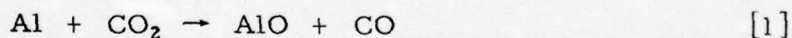
The agreement between (extrapolated) rate coefficients obtained from high and low temperature determinations by separate techniques is often poor. It is also becoming apparent that, for many reactions, Arrhenius-type

plots of rate coefficients vs. T covering ranges on the order of 1000 K or more show distinct upward curvature with increasing T (e.g. Refs. 2, 3), thus making extrapolation of $k(T)$ data over wide temperature ranges a procedure of doubtful validity. For reliable $k(T)$ measurements it is desirable to use a single technique to span the entire T -range of interest. For the 300-2000 K range our high-temperature fast-flow reactor (HTFFR) technique provides such a technique for the first time.

Discussion

In the report period we concentrated on checking and elaborating on results obtained earlier in the contract year (4), during which we had performed studies on the Al/CO_2 and $\text{Al}/\text{H}_2\text{O}$ reactions.

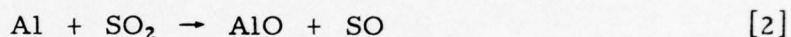
The reaction



has a $k_1(T)$ which increases from 700 to 1800 K much more rapidly than the simple Arrhenius equation would suggest (4). To check if CO_2 dissociation and the resulting O_2 production could have occurred to a sufficient degree to influence the measurement of k_1 (1800 K) we calculated the extent of equilibrium dissociation (which is probably not achieved due to the short reaction times in the HTFFR). These results were then used to re-analyze the kinetic data. The results indicate that the effect of such dissociation is well within

experimental error. Some mass spectrometric checks on the results of these calculations were attempted. However, the available mass spectrometer was found to be inadequate for this work. The instrument has now been improved and further checks are about to be made.

Over the 300 to 700 K range k_1 (T) was found (4) to behave according to the Arrhenius law, which allows determination of an activation energy, found to be $2.3^{+1.3}_{-0.7}$ kcal mole⁻¹. This value implies $D(\text{Al-O}) \geq 123.7^{+0.7}_{-1.3}$ kcal mole⁻¹. The lower limit value of 122.4 is equal to the maximum value allowed by JANAF (5) and casts some doubt on this accepted $D(\text{Al-O})$. Independent of the SQUID contract, we have investigated the reaction



at one temperature (700 K) which suggests that $D(\text{Al-O})$ is at least 5 kcal mole⁻¹ higher than the JANAF figure. (Reaction [2] was chosen since the SO-O bond, ≈ 131 kcal mole⁻¹, is ≈ 5 kcal mole⁻¹ stronger than the CO-O bond and hence Reaction [2] has a $\Delta H \approx 5$ kcal mole⁻¹ higher than Reaction [1]). This larger value of $D(\text{Al-O})$ would be in agreement with the flame photometric work of Jensen and Jones (6) which indicates a value of 132 ± 6 kcal mole⁻¹ (7) when corrected for the newer more reliable f number of the $\text{AlO}(\text{B-X})$ transition (8). Since an increase in the accepted (5) value of $D(\text{Al-O})$ would have important consequences for prediction and modelling of the exhausts of rockets using aluminized propellants, we are currently studying the Al/SO_2 reaction over a range of temperatures (300-700 K initially) to arrive at an accurate upper limit for the activation energy of

Reaction [2] and hence a lower limit for $D(\text{Al-O})$.

Further investigation of the $\text{Al}/\text{H}_2\text{O}$ reaction at ≈ 600 K revealed a strong dependence of the apparent rate coefficient on flow velocity, suggesting that under the conditions investigated the Al-consumption is dominated by wall reactions. While it still may be possible, under other experimental conditions, to obtain homogeneous $k(T)$ for this reaction the effort involved appears unwarranted in view of the limited budget. We have therefore temporarily put this study aside in favor of the above-mentioned $D(\text{Al-O})$ work and the planned investigations of the Al/HCl and AlO/CO_2 reactions.

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EXPERIMENTAL AND THEORETICAL STUDIES OF MOLECULAR COLLISIONS AND CHEMICAL INSTABILITIES

Massachusetts Institute of Technology, Cambridge, Massachusetts
Subcontract No. 4965-10

Professor John Ross, Chief Investigator
Dr. George Schatz
Mr. Randolph Burton

Introduction

The research program is concerned with theoretical and experimental studies of molecular collisions in reactive and non-reactive systems, and theoretical and experimental studies of chemical instabilities.

Discussion

A. Chemical Instabilities. The experimental work on the observation of chemical hysteresis, multiple stationary states, and increase in fluctuations near marginal stability in the simple chemical reaction $2\text{NO}_2 = \text{N}_2\text{O}_4$ under illumination by an argon ion laser has been written up as an article and accepted for publication in the Journal of Chemical Physics.

We have made further progress in the study of statistical (stochastic) theories of the kinetics of nucleation in phase transitions. We begin with a postulated master equation for the variable of interest such as density, and derive by path integral methods the deterministic equations for the most probable behavior of the system. If, in this process, we impose a constraint of local conservation of the variable of interest then we obtain the Cahn equation which is of the diffusion type. The same derivation without this constraint has been shown previously to lead to a Landau-Ginzburg equation which in addition to a diffusion term has a source term in it. We have shown that the two equations have a common origin. The Cahn equation is applicable to conserved variables such as numbers of particles in systems in which spatial inhomogeneities occur over distances much larger than a characteristic distance λ of the system (such as mean free path or nearest neighbor separation). The Landau-Ginzburg equation is applicable to nonconserved variables (such as magnetic spin) and to conserved variables when spatial inhomogeneities occur over distances much smaller than the characteristic distances for those variables (for instance the size of a droplet in a gas undergoing nucleation to form a liquid). The derivation presents an understanding of the relation of these two types of equations and shows the limits of applicability of each. An article has been written and published in the Journal of Chemical Physics.

We have studied cooperative effects in energy relaxation and energy transfer for a system of N atoms coupled to a common radiation field.

In such systems, in addition to these cooperative effects super radiance may occur and this process has some properties analogous to instability phenomena. We show that the system may to some approximation be described by nonlinear rate equations and further show that energy transfer processes within the system may have oscillatory time behavior. Locally excited states show cooperative behavior in their decay. Analysis of a form of the superradiance master equation for spatially large systems reveals the existence of two elementary cooperative processes whose significance decreases with increasing distance between the atoms involved: one is the transfer of energy through the field between two atoms; the other is a cooperative loss of two quanta of energy to the field by two separate atoms. An article on this work has been written and accepted for publication in *Physica*.

B. Chemical Dynamics. For the reaction $\text{Ba} + \text{Cl}_2$ we have made molecular beam measurements of one reaction channel in which the products are $\text{BaCl}^+ + \text{Cl}^-$ against another reaction channel in which the product is BaCl_2 and a photon. The BaCl_2 molecule is formed by a collision which is then stabilized by a subsequent collision. Part of this work has been written up for publication.

We have constructed and have tested a molecular beam source for ozone and are now in the process of using this ozone source to study oxygen atom transfer reactions such as $\text{O}_3 + \text{NO}$ and others.

We have derived and shown the utility of an approximate theory of chemical dynamics based on a generalized Franck-Condon factor. We begin by showing how the general expression for the transition matrix

for an electronically adiabatic reaction may be rewritten in terms of a transition between two surfaces through the use of a quasiadiabatic representation. This exact transition matrix may be reduced to a Franck-Condon overlap integral in a variety of ways, and one possible sequence of approximations for accomplishing this reduction is outlined. We neglect terms due to virtual transitions to excited electronic states, make a Born-Oppenheimer approximation, neglect terms involving gradients of the nuclear wavefunction (low kinetic energy approximation), and finally make a Franck-Condon approximation.

The overlap is then evaluated for the special case of collinear exoergic atom-diatom reactions for the purpose of studying product state vibrational distributions in these reactions. The evaluation is done approximately by using physical arguments to estimate the general appearance of the reagent and product quasiadiabatic surfaces, and assuming separable solutions to the Schrödinger equation on each surface. The overlap integral is then further approximated by expanding the integrand about the nuclear configuration of maximum overlap. This enables us to obtain a simple analytical result for the product state distribution, using either harmonic or Morse oscillator vibrational wavefunctions. We then use the resulting expressions to study the dynamics of the collinear $F + H_2(D_2)$ and $H(D) + Cl_2$ reactions. In both applications we find that the Franck-Condon overlap is capable of a qualitatively correct description of the product state distributions, including dependence on reagent translational energy, mass ratios and various features of the potential energy surface. Furthermore, a

physical description of the origin of a dynamic threshold effect in the $F + H_2(D_2)$ reaction is provided, as is a simple interpretation of the role of potential energy release behavior in the determination of product state distributions.

We have also studied vibration-rotation distributions in two and three-dimensional atom-diatom chemical reactions using the generalized Franck-Condon overlap theory of chemical dynamics. We begin by reducing the Franck-Condon overlap form of the transition amplitude to a partial wave series involving overlap expressions for the partial wave transition matrices. These quantities are then approximately separated into products of angular coordinate factors and vibration-translation factors. To evaluate the angular coordinate factor, we examine the limits of strong and weak potential and kinematic coupling, where simple dynamic approximations (such as sudden approximations) may be used, and we are thus able to do the integrations analytically. The vibration-translation factor is evaluated by reducing it to an overlap similar in form to one previously analyzed for collinear reactions (Ref. 1), with some differences arising from centrifugal forces.

The resulting formulas for product internal state distributions are then applied to the 2D and 3D $H+H_2$ reaction, and to the 3D exoergic reactions $F + H_2(D_2)$ and $H(D) + Cl_2$. For $H + H_2$ we find that centrifugal effects may be ignored, and that the angular coordinate part of the Franck-Condon overlap provides an accurate qualitative description of the rotational distributions. In addition we find that on reaction the projection quantum number distribution shows a marked propensity

for the condition $m_j = m_{j'} = 0$ for the z component of the rotational angular momentum. Centrifugal effects are important for the exoergic reactions considered, and the simple formulas developed for describing vibration-rotation distributions for such reactions give a qualitatively (and often quantitatively) accurate description of the shapes of these distributions and their dependence on dynamical and kinematic parameters such as mass ratios, potential anisotropy and energy release behavior. In addition, a qualitative understanding of the Levine-Bernstein information theoretic procedure for relating 1D and 3D vibrational distributions is provided, and possible sources of errors in this method are identified.

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2. "Nonequilibrium Kinetics: Exact and Approximate Solutions," J. Stat. Phys. 14, 469 (1976). S. Hudson and J. Ross.
3. "Superradiance and Energy Transfer Within a System of Atoms" Accepted for publication in Physica. J. P. Stone, A. Nitzan and J. Ross.
4. "Dynamical theory of migration of an adsorbed atom on solid surfaces," accepted for publication in J. Chem. Phys. K. Kitahara, H. Metiu, R. Silbey, and J. Ross.
5. "Multiple stationary states and hysteresis in a chemical reaction," accepted for publication in J. Chem. Phys. C. L. Creel and J. Ross.

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9. "Franck-Condon factors in studies of dynamics of chemical reactions II. Vibration-rotation distributions in atom-diatom reactions," submitted to J. Chem. Phys. G. C. Schatz and J. Ross

Lectures

The Principal Investigator presented invited lectures at:

American Physical Society, Symposium on Instabilities.

Max Planck Institut for Biophysical Chemistry, Göttingen.

Annual Meeting of Bunsengesellschaft Plenary Lecture.

Gordon Conference on Dynamical Instabilities.

Gordon Conference on Chemical Dynamics.

A BASIC STUDY ON THE MECHANISM OF INFLAMMABILITY LIMITS
AND THE BEHAVIOUR OF NEAR-LIMIT FLAMES

Case Western Reserve University
Cleveland, Ohio
Subcontract No. 8960-2

Professor James S. T'ien, Principal Investigator

Introduction

The research program is concerned with the mechanism of flammability. Both steady and unsteady considerations are included in order to explain the behaviours of flames near the limits.

Discussions

Experiments were conducted to study the spontaneous flame oscillation phenomena before the flame goes to extinction. Near-limit flame oscillation has been observed by people performing limiting oxygen index tests^(1,2). But their descriptions of the phenomena were restricted to one or two sentences and no systematic pursuit of this aspect has been made. Recently, two theoretical studies of flame stability, one involved a gaseous diffusion flame⁽³⁾ and the other on solid propellant low pressure extinction limit⁽⁴⁾, suggested that the extinction limit of flames might be a dynamic neutral stable point with natural frequency other than zero. In the unstable domain, a flame will oscillate spontaneously with increasing amplitude. Based on observations of solid rocket combustion with L-star type instability, it was postulated in Reference 4 that this kind of

oscillation will lead to dynamic extinction when the oscillating amplitude becomes too large. These theoretical results led to the present experimental investigation.

Type of fuels tested included kerosene, heptane, ethanol, methanol, commercial candles, plexiglass and polyethylene rods. The first type of tests were done in a large combustion chamber (24"x17"x16"). All liquid fuels and candles were burned with wicks, plexiglass and polyethylene rods were burned from the top end with or without a container. The flames were ignited in atmosphere, then the chamber pressure was reduced to a level close to the low pressure extinction limit. The flame was left to burn by itself. As the oxygen in the combustion chamber was gradually consumed, extinction limit was approached. This quiet and gradual approach of the limit was needed to eliminate external disturbances and to insure good observation of the spontaneous oscillation that we are looking for. The tests showed that plexiglass rod flame exhibited no visible oscillation. Flame from polyethylene rod without container vibrated in a irregular wobbly manner probably because of the melts on the side of the rod created an unsymmetric configuration; and this vibration is not restricted to near-limit regime. Flame of polyethylene with a container, candle flame and all liquid wick flames exhibited spontaneous oscillation starting from a steady state. As can be seen from the attached photograph of candle flame, the luminous region of the flame vibrated in size. The oscillating amplitude increased in time and finally flame extincted in a dynamic manner. The oscillating fre-

quencies varied from 6 to 10 Hertz depending on the type of fuels and burner set-up. The oscillation could last from a few cycles to a couple of dozen cycles before extinction depended mostly on the type of burners. Since all experiments of this type were done at low pressure (2 to 7 psia), flames were mostly blue in color with the yellow soot zone either eliminated or greatly reduced in size. As a consequence, the flame appearance was quite different from that of the atmospheric flame as it can be seen in the attached photograph.

The second set of tests were done in small diameter tubes (7/16" and 7/32") for liquid fuels only. The tube was first filled with liquid and then ignited. As the fuel was gradually consumed, the liquid surface level receded. Depending on the types of fuels and the tube diameter, the flame either stayed at the opening of the tube or went into the tube. In the first case, flame was quenched because fuel vapor supply was reduced when the liquid surface became too distant from the flame. In the second case, the oxygen supply became increasingly difficult as the flame itself went into the tube. Heat loss to the tube wall also contributed to quenching in both cases. These tests have been done both at atmospheric and lower pressures. Oscillation before extinction is always observed although it usually lasts only a few cycles before extinction.

In order to understand the mechanism of near-limit oscillations, simultaneous temperature measurement at different locations in the flame field is being made to determine what type of time lag that may exist. In addition, temperature measurement just before the

oscillation was to begin indicates that near the bottom of flame, temperature was about 300°F lower than that at the flame top. Since the oscillation occurs in a way that the bottom edge of the flame goes up and down but the flame top remains pretty steady (see figure), it suggests that the oscillation may be a local extinction and re-ignition event. This would imply that the phenomena is two or three dimensional in nature, the one-dimensional analysis in References 3 and 4 will be inadequate. More experimental data on oscillating temperature is being analyzed before we can draw a final conclusion.

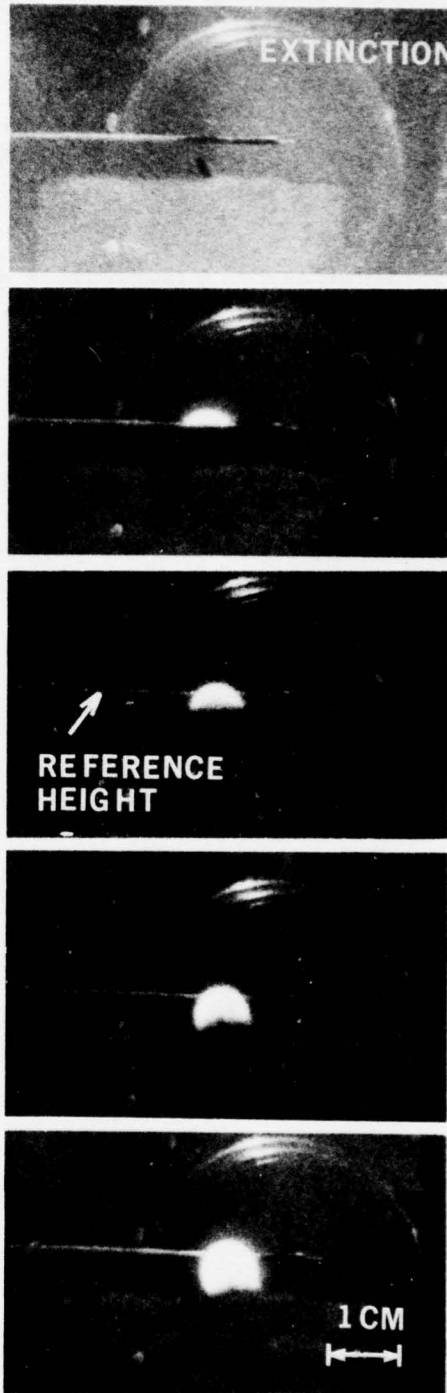
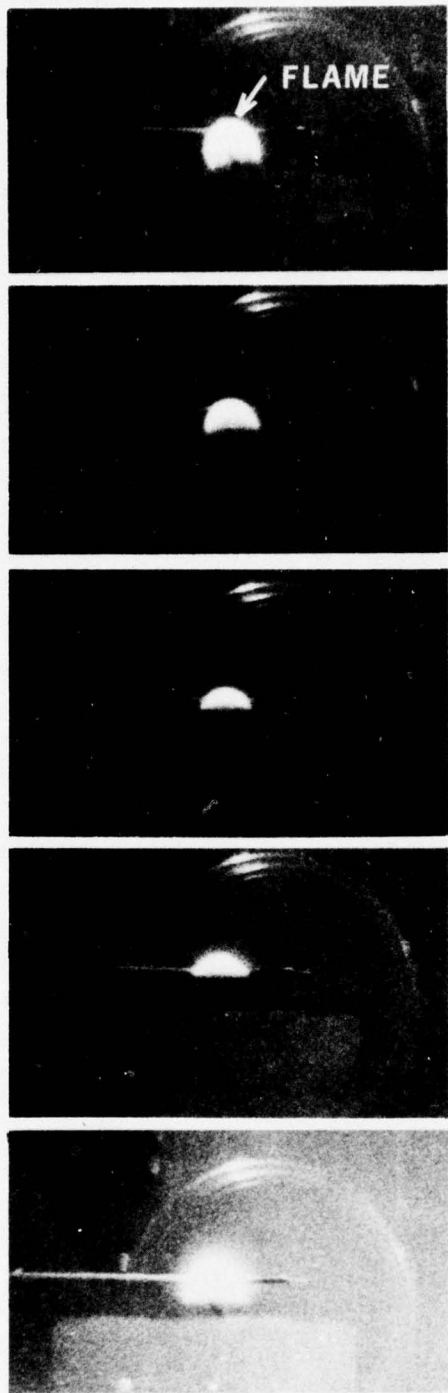
It should be emphasized that the oscillation we discussed here is a near-limit phenomena. This is different from flame oscillations⁽⁵⁻⁸⁾ previously found which were attributed to hydrodynamic instability in the flame jet and have nothing to do with the extinction limit.

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DIRECTION OF EXPOSURE
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NEAR-LIMIT OSCILLATING CANDLE FLAME AT 2 PSIA. FILM SPEED IS
32 FRAMES PER SECOND.

III. MEASUREMENTS

INVESTIGATION TO EXTEND THE APPLICABILITY OF LASER RAMAN
SCATTERING DIAGNOSTIC TECHNIQUES TO PRACTICAL COMBUSTION SYSTEMS

United Technologies Research Center
East Hartford, Connecticut 06108
Contract 8960-20

Alan C. Eckbreth, Principal Investigator

Introduction

With the development of high power, visible laser sources, Raman scattering techniques have received considerable attention for combustion diagnostics in the past several years. Laser Raman approaches have been advanced to the point where they are now being routinely employed in a variety of fundamental flame investigations. However, widespread application of laser Raman techniques to practical combustion devices such as gas turbines, hydrocarbon combustors, furnaces, etc., is yet to be achieved. From an instrumentation standpoint, practical devices contain flames which differ markedly from those typically employed in fundamental laboratory studies. Environments of practical interest contain flames which are generally turbulent, high luminous and particulate laden. The high luminosity levels preclude the use of cw laser sources for Raman work in these environments. Pulsed laser sources are required to produce peak Raman powers comparable to or preferably in excess of the background luminous power. In some instances, low S/N ratios are encountered even with pulsed lasers. In situations where the background luminosity is tolerable, the pulsed laser-particulate interaction can lead to a

variety of laser-particulate interaction "noise" effects which can adversely affect the successful application of laser Raman techniques. In laser Raman thermometry experiments at UTRC, successful Raman measurements in a hydrocarbon-fueled model combustor were, in one case, precluded by laser modulated particulate incandescence. Particles (e.g., soot) in the measurement volume, which at high stream temperatures are already incandescent and contribute significantly to the total luminous emission, absorb the incident Raman-inducing laser radiation, heat to temperatures far above ambient, and emit greatly increased amounts of gray/blackbody radiation which can exceed the sought-for Raman signal levels. Under Project SQUID sponsorship, this problem and potential solutions have been experimentally investigated under controlled laboratory conditions. Details of these studies will soon be available in a Project SQUID technical report (Ref. 1).

Discussion

Detailed analyses (Ref. 2) of laser irradiated particle heating have been made in an effort to obtain a better physical understanding of this phenomena and to seek solutions to laser induced or modulated incandescence "noise" effects. In general, these analyses display the inadequacy of the various heat transfer processes from preventing the surface temperature from rising appreciably above the initial level, particularly for larger particle sizes, i.e., $\geq 1\mu$, and higher focal flux values, i.e., above 10^5 W/cm^2 . Due to this inadequacy, the particle surface temperature tends to follow the laser pulse envelope exhibiting little phase behavior which could be exploited for noise suppression. Despite the high

surface temperature produced, the analysis predicts a relatively weak dependence of surface temperature on focal flux. Thus, even though the surface temperature and the incandescence rises with increasing focal flux, the incandescent noise does not increase as rapidly as the Raman signal, and S/N improvements are predicted with increasing levels of focal flux.

Experimental Laser Irradiated Particle Noise Studies This aspect of the investigation addressed the validity of foregoing analysis. Initially laser induced particulate noise was studied by irradiating singly suspended 5μ diameter particles. Early results, including the presence of laser induced C_2 Swan emission in addition to incandescence, were summarized previously (Ref. 3). One important point worth reemphasizing concerns the distinction between alumina and carbon (soot) particulates which has been experimentally verified. Unlike soot particles, Al_2O_3 possesses an extremely low absorption cross section, which varies from four to five orders of magnitude less than its geometric cross section (Ref. 4) depending on particle size and wavelength. Consequently, very little incident laser energy is adsorbed by an alumina particle resulting in very little heating and, hence, very little laser induced radiative noise. Hence, Al_2O_3 would be an excellent seed for LDV work permitting simultaneous LDV and laser Raman studies.

Particle size measurements in the UTRC test combustor (Ref. 5) using a Mie scattering technique indicated an average particle size of 400 \AA (0.04μ). Consequently, subsequent testing focussed on laser induced particulate noise produced by laminar propane diffusion flames whose soot sizes were known to be in this range. Laser induced particulate radiation was monitored as a function of energy

and focal flux employing a two channel Raman spectrometer (Ref. 2). For these studies, the spectrometer functioned as a two color pyrometer (5230, 6864 Å) and was calibrated against a tungsten filament as known temperature. Laser heated particulate temperatures, determined in the above manner, as a function of focal flux level exhibited fairly good agreement with the analytical model (Ref. 2). The particulate temperatures varied from approximately 3500°K to 4500°K over a flux range from 10^5 to 10^8 W/cm². Quite importantly, the absolute noise level after increasing initially with laser energy, leveled off and became nearly independent of energy for laser energies in excess of about 60 mJ. This results essentially from the saturable behavior of the laser heated particle surface temperature with increasing focal flux. The laser induced particulate noise also decreased with decreasing focal length due to the decrease in the number of particles being irradiated. From this data, a signal to noise improvement of several hundred was extrapolated for flux increases from $2(10^5)$ W/cm² to 10^9 W/cm². Operation beyond this flux level may be problematical due to gas breakdown. These results clearly indicate the value of operating at the highest possible focal flux level in particulate laden environments. Operation at these flux levels can lead to other problems, however, such as optical damage in and fluorescence from window ports, both of which have been experimentally encountered.

Signal Averaging Considerations In Ref. 6, the consequences of time averaging Raman data generated by a cw laser from a turbulent medium were examined. In an analogous fashion, an analysis of ensemble averaging pulsed Raman data in a fluctuating medium was performed. The analysis can handle such effects as laser

energy variations and shot noise fluctuations and treats among other items, the case where "noise" is sampled and subtracted from a "signal" channel. The analysis demonstrated that when Raman data are averaged, background can be subtracted on average and shot noise terms average to zero. However, temperature inferred from the ratio of separately average band intensities will not be the average temperature but will depart from the true average depending on the magnitude and correlation of the fluctuations. Furthermore, in measurement situations where the band intensity ratio at any instant cannot be determined accurately (due to shot noise effects, including those arising during subtraction), the average of these separate erroneous temperature measurements will not yield the true average temperature.

Seeded Flame Temperature Measurements A CH_4 diffusion flame sustained in the center of a 7.6 cm dia. premixed flat flame burner was employed to introduce controlled amounts of laser induced particulate noise into a Raman temperature measurement experiment. A four channel Raman spectrometer consisting of two signal and two noise channels, the latter located in spectral regions just adjacent to the Raman bands, was used to demonstrate noise sampling and subtraction. Due to shot noise fluctuations, good subtraction is not obtainable with each pulse, but only on average over a large number of pulses depending on the photon levels involved. By adjusting the noise channel photomultipliers in ten volt increments good subtraction was obtained on average with the dye laser so tuned that all four channels "saw" only noise. By appropriate tuning of the laser, the Raman bands were placed within the signal bandpasses and observed with the noise subtracted

out. These signals were compared with Raman data with the seed flame extinguished and agreed quite well since the introduction of the seed flame constituted only a small perturbation on the burner temperature profile. Thus noise sampling and subtraction is a viable approach for Raman measurements in noisy media subject to potential errors arising from signal averaging in unsteady environments.

Conclusions and Recommendations It is difficult to be categorical in regard to the utility of spontaneous Raman scattering for practical combustion diagnostics since conditions can vary widely from device to device and within a given device depending on location and operating parameters. Based upon our studies, primary zone diagnostics in a diffusion flame apparatus operating on hydrocarbon fuels appears highly doubtful based upon both background luminosity considerations and laser induced particulate noise effects. Secondary and exhaust region probing may be possible (Ref. 7) since luminosity levels tend to be lower, but particulate effects could be problematical if the particles are in sufficient quantity or size. High focal flux levels appear optimal for enhancing Raman S/N in regard to laser induced particulate noise but may lead to problems with window breakdown and fluorescence. The latter may be sufficiently strong to preclude the use of preferred background luminosity suppression geometries. Noise sampling and subtraction is feasible as has been demonstrated. However, shot noise effects dictate that Raman data be separately averaged to permit accurate determination of average signal levels. Ensemble averaging, however, leads to band intensity ratios which, in fluctuating environments, depend not only on average temperature,

but on average density and on the magnitude and correlations of fluctuations in density and temperature. These effects may result in large measurement errors rendering the extracted Raman data of little utility. Based on this perception of the situation, we believe spontaneous Raman scattering to be of limited utility for many practical applications. However, due to its relative simplicity and high level of present day understanding it certainly merits first consideration in environments where the aforementioned noise effects are not overwhelming.

For most practical applications, the feasibility of stronger Raman processes such as CARS (coherent anti-Stokes Raman scattering) or near-resonant Raman scattering needs to be investigated. In particular CARS has shown great potential to date but is not without problems due to its sophistication. It is not completely clear at this point what the impact of practical device characteristics such as high turbulence and particulate levels will be on CARS generation. In particular since nonresonant susceptibility contributions are often problematical in CARS experiments, potential particulate (soot) noise effects should be examined in a controlled manner.

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TEMPERATURE, CONCENTRATION, VELOCITY AND
TURBULENCE MEASUREMENTS IN JETS

Polytechnic Institute of New York

Subcontract No. 8960-5

S. Lederman - Principal Investigator

Introduction

As indicated in the last semi-annual progress report of March 1976, in order to take advantage of the thus far developed non-intrusive diagnostic techniques, and utilize the same to their full capabilities, an expansion of the data acquisition system was undertaken. The introduction of additional data acquisition channels and the expansion of the memory bank of our on line computer has been completed. Additional expansion of our data acquisition and processing system is now under way with the ultimate aim of providing a complete diagnostic system capable of providing the experimental answers to the problems of temperature, concentration, velocity, turbulence measurements in flowfields, high and low pressure combustion systems as well as in exhausts of internal combustors.

Discussion

The high power laser pulse technique as indicated in Ref. 5, 6 and 7 is capable of providing instantaneous and simultaneous in-

formation not only concerning temperature and concentration of a single specie in a flow field, but also, depending on the number of photoelectronic data acquisition channels, and data handling capabilities of the information gathering, storage and processing system available, information concerning concentration, temperature and accuracy of as many species as may be present in the flow field, provided of course, that the species are Raman active. As pointed out in Ref. 5, the velocity field and turbulent intensity may be obtained simultaneously. From previous discussions Ref. 1, 5 and 7 it is clear that the acquisition of the temperature of a single specie requires essentially 3 data acquisition channels operating simultaneously. One of these channels which may be used subsequently for the whole system is being used as a laser power reference channel. The other 2 channels provide the information concerning the stokes and anti-stokes Raman intensity for the given specie of interest. The ratio of the last 2 provides the temperature, and the stokes and anti-stokes intensity by themselves, provide the concentration. Thus a system containing for example only 3 species, requires as much as 7 data acquisition photoelectronic storage and processing channels. If any additional information concerning velocity and turbulence intensity is required, an additional channel must be provided. At the time of the last progress report our system has been expanded to 4 channels. Since then, additional 3 channels have been installed and are now being calibrated.

It has been discussed (Ref. 7) previously that under certain

conditions the acquisition of data concerning concentration and temperature may be impossible using the spontaneous Raman effect. This may be particularly true in certain cases of combustion where the concentration of the specie of interest may be very low, or where fluorescence and other sources of noise may exceed the radiation intensity of interest. In this case, the application of the Coherent Anti-stokes Raman scattering technique (CARS) may be useful. At the time of the last progress report, a preliminary result concerning hydrogen concentration using the CARS method has been presented. At this time, a similar curve concerning methane is shown in Fig. 1. In addition, some data have been obtained using CARS in a turbulent flame. These data while definitely proving the applicability of CARS to combustion diagnostic are at the moment only of a qualitative nature, and, therefore, not suitable for presentation.

At this point it is worthwhile mentioning that the CARS system presently used in our laboratory consists of a Ruby laser, a stimulated Raman cell in which the specie of interest is being used under elevated pressure to produce the Stokes line, which is then combined with the Ruby line in the flow field to produce the anti-stokes line of the specie of interest.

As is well known, the production of the stimulated Raman emission of some species may present some difficulties. For example, the generation of the Stokes line from water vapour by means of stimulated Raman emission is quite difficult for obvious reasons.

To avoid difficulties of this kind, a tunable dye laser would be desirable. At this time a complete system consisting of an argon ion laser, a tunable dye laser, and an appropriate data acquisition apparatus has been placed on order. This equipment should be available in the laboratory by the 1st of October, 1976.

Presently work is proceeding concerning the acquisition of temperature, concentration, velocity and turbulence intensity in jets and flames. Furthermore, the extraction of turbulence data from the Raman obtained temperature and concentration is receiving considerable attention.

Some of the data obtained are in the process of being evaluated and should be soon available in the form of a report. Fig. 2 and 3 represent some of the raw data obtained simultaneously using Raman and L.D.V. respectively on a flame.

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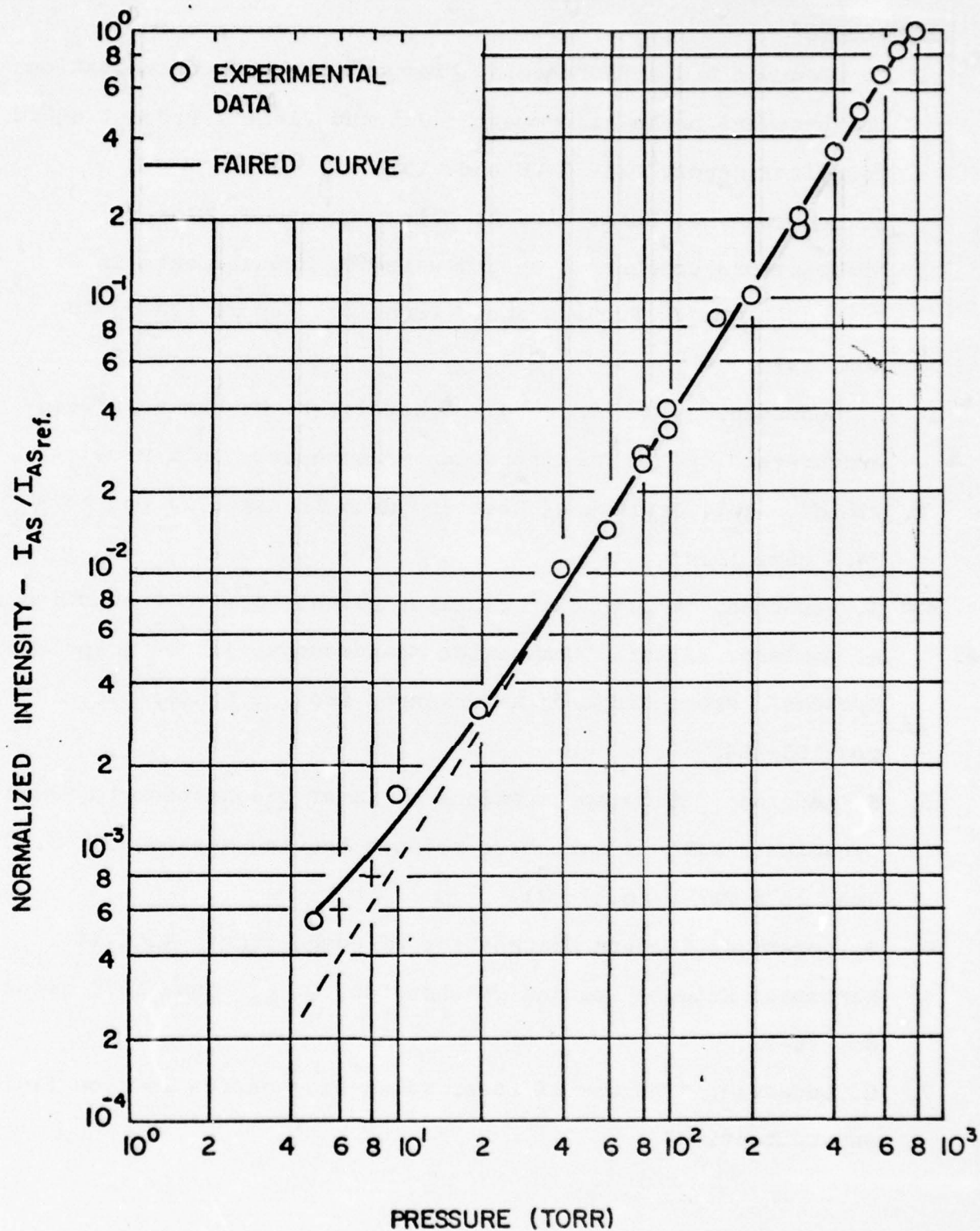


FIG. 1 MEASURED COHERENT RAMAN ANTI-STOKES INTENSITY OF METHANE (CH_4) AS A FUNCTION OF PRESSURE

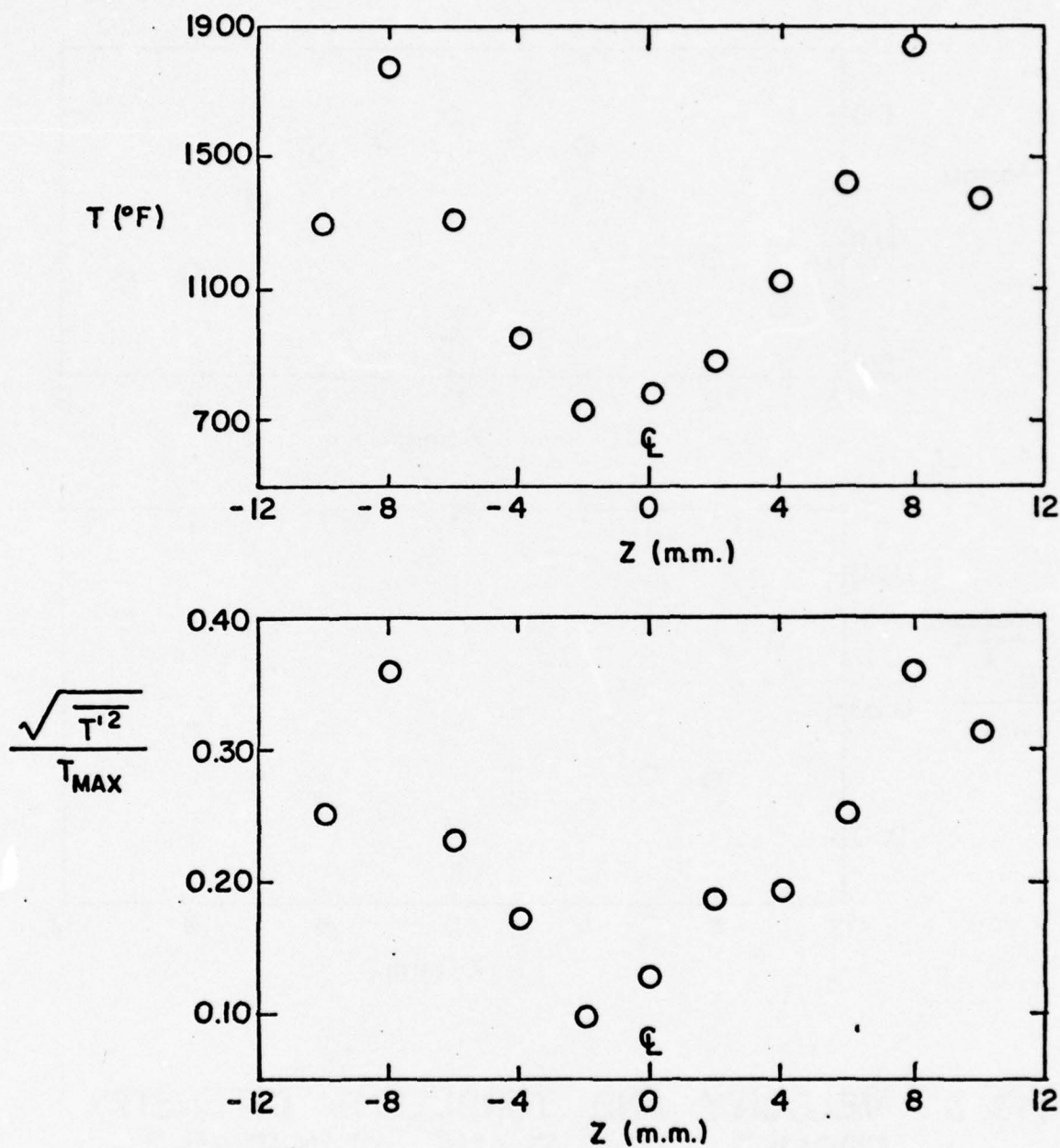


FIG. 2 TEMPERATURE AND TURBULENT INTENSITY
IN A FLAME AT $X/D = 5.2$

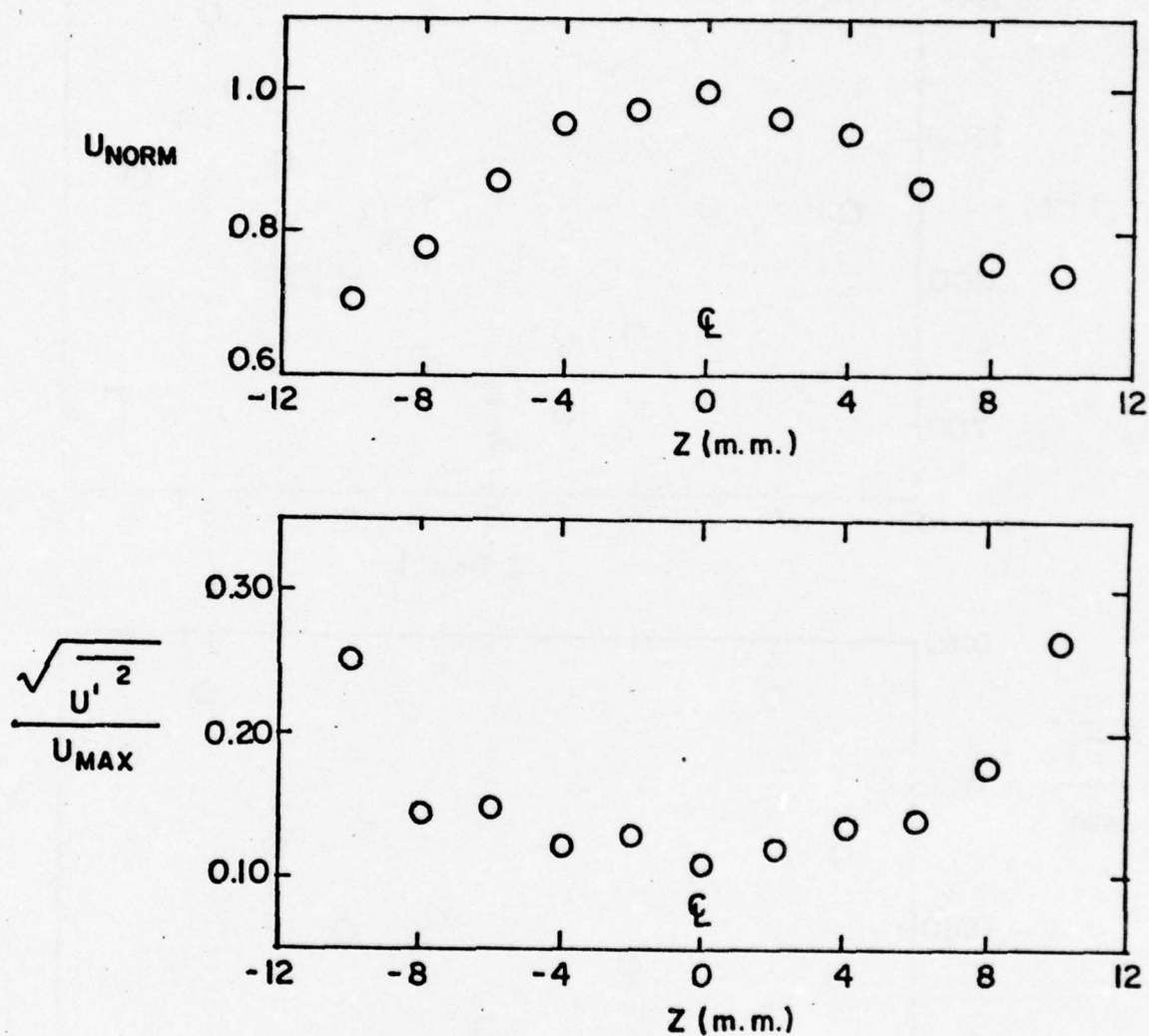


FIG. 3 VELOCITY AND TURBULENT INTENSITY PROFILE IN A FLAME AT $X/D = 5.2$

LASER RAMAN PROBE FOR COMBUSTION DIAGNOSTICS

General Electric Company, Corporate Research and Development
Schenectady, N. Y.
Subcontract No. 8960-17

Marshall Lapp, Principal Investigator
James C.F. Wang, Co-Investigator
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C.M. Penney, Physicist

Introduction

Axial velocity histograms have been obtained for a hydrogen-air flame produced inside a coaxial jet burner, through the use of a laser velocimeter. Flames at two hydrogen-to-air speed ratios were tested in this program. A comparison of the data with and without combustion demonstrates that both mean velocity and turbulence level increase with combustion. The increase in mean velocity peaked along the axis of the hydrogen jet, while the increase in turbulence peaked at the mixing region of the hydrogen-air coaxial jet flow. The explanation for these mean velocity and turbulence changes will become clearer with further planned experimentation, including vibrational Raman scattering measurements of instantaneous values of temperature and density for this same flame system.

Discussion

The objective of our Project SQUID program for 1975-76 was two-fold: (1) to demonstrate the capability of the laser velocimeter (LV) technique for flow velocity measurement in a combustion system, as a first step toward implementation of simultaneous measurements of velocity with LV and temperature, density, and major species composition with vibrational Raman scattering; and (2) to provide useful information on flow turbulence with combustion in a well-defined flow environment.

A well-defined and controlled hydrogen flame was obtained in a coaxial hydrogen-air burner consisting of a 2.7 mm-diameter hydrogen jet on the axis of a surrounding air pipe. The air flow from a blower was distributed, straightened, and developed into a turbulent pipe flow in a 100-mm-diameter smooth copper pipe. The test section of the burner was made from a glass pipe of the same diameter and the hydrogen jet was translated along the burner centerline to provide velocity measurements at various downstream stations without moving the LV optical system. At each downstream station, the radial velocity profile was measured by manually moving the LV transmitting lens perpendicular to the burner centerline. The LV optical system for these experiments was a dual scatter device utilized in an off-axis backscattering arrangement. The diameter of the scattering volume was about 0.3 mm and the length about 0.5 mm, with a fringe spacing of 3.1 μm . A direct frequency counter LV processor developed at GE CRD was used to acquire and validate the LV data. Mean and turbulent velocities were obtained from the histograms from a pulse

height analyzer used in conjunction with the LV processor. Alumina powder of 1 μm nominal diameter was used as the seeding material for these LV measurements. A strong luminosity in the flame (probably from Na impurities in the alumina) was rejected through the use of an optical filter in front of the LV photodetector.

Reasonably axisymmetric and fully developed pipe flow profiles have been obtained for the coaxial air flow surrounding the hydrogen jet at a mean velocity of 14.5 m/s, with the hydrogen and air at room temperature ($\approx 21^\circ\text{C}$). A comparison of the LV data with pitot-tube measurements was performed with only air flowing in the burner. The averaging effect caused by the large physical size of the pitot tube was observed clearly in the wake region of the hydrogen jet tube (which was, itself, comparable in diameter to the pitot tube and about 10 times larger than the LV scattering volume). Outside the wake region, LV and pitot measurements agreed within $\pm 1\%$ accuracy.

LV measurements were performed on the coaxial hydrogen-air flow both with and without combustion in order to study the effect of burning on flow turbulence. Two flame conditions were tested at hydrogen-to-air velocity ratios of approximately 6:1 and 3:1, with qualitatively similar results. The mean and turbulent velocities measured without combustion but with hydrogen flow were used as the base line and subtracted from those obtained with combustion at the same flow conditions. The resultant mean and

turbulent velocities directly represent the effects of the combustion process, and therefore we discuss our results in terms of incremental quantities.

Based on the LV data from two speed ratio conditions, increases in both mean ($\Delta \bar{U}$) and root mean square turbulent axial velocities $\Delta \sqrt{u'^2}$ were found due to the presence of combustion. The increase in mean velocity was peaked at the burner centerline, which is also the axis of the hydrogen jet, at each downstream station. The increase in mean velocity due to combustion along the burner centerline was found to decrease with distance from the hydrogen jet exit, starting with observations made at 50 jet diameters downstream; the maximum increase in this mean velocity occurred at an axial position of 50 jet diameters or less downstream.

The radial profiles of the axial root mean square turbulent velocity increase were different from the mean velocity increase radial profiles, and depended on the axial location with respect to the closing of the visible flame envelope on the centerline of the burner. Within the visible flame envelope, the radial profile of $\Delta \sqrt{u'^2}$ had two peaks that were symmetric about the centerline and located radially inside the visible flame. Measurements made at two axial locations inside the flame envelope indicated that the peak magnitudes of $\Delta \sqrt{u'^2}$ were the same.

Downstream of the visible flame envelope, we observed a single maximum in the radial profile of $\Delta \sqrt{u'^2}$, which was of a rough plateau shape. The peak magnitude was less than that for the peaks observed inside the flame envelope, and was found to decrease with axial distance downstream.

The experimental results indicated that both the mean velocity and the root mean square turbulent velocity increased substantially in the presence of combustion. The increase in turbulence level can be attributed to the increase in mean flow velocity as well as to direct coupling from the combustion process, e.g., small scale eddy dilation, heat release, etc. We are presently completing preliminary thermocouple measurements to obtain mean temperature profiles at corresponding LV velocity profile locations. Further interpretation of these turbulence data will need more detailed complementary measurements of instantaneous values of temperature. An experiment utilizing vibrational Raman scattering to produce this necessary information is currently under development.

Notes and References

Recent publications and manuscripts related to this research effort supported by Project SQUID and by other parallel General Electric and government efforts are listed below:

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INVESTIGATION OF NOVEL LASER ANEMOMETER
AND PARTICLE-SIZING INSTRUMENT

1 April 1976 to 30 September 1976

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Initially it was proposed to investigate a dual focus transit-timing anemometer first, and to follow this by an investigation of methods for in situ particle sizing, simultaneous with velocity determination on individual particles. However, in view of the more urgent need for a method of particle sizing, it was decided to reverse the priorities and tackle the more difficult particle-sizing aspect first.

Accordingly, in this first year, attention has been focused on developing a light scattering method for in situ sizing of individual particles. This was summarized in the semi-annual report and an abbreviated account has been published (1) as part of an invited review on "Diagnostic Methods in Combustion MHD Flows" under another contract.

As a result of this survey, a particular concept was adopted for investigation, which should be suitable for in situ sizing of particles in the range 2-50 μm or more. This concept is based on collecting the whole of the forward diffraction lobe scattered by a particle

traversing a line focus produced from a laser beam by a cylindrical lens (Figure 1). The forward diffraction lobe yields a strong, well-characterized scattered signal which, for particles greater than $\sim 2 \mu\text{m}$ with visible light, is proportional to particle projected area, is insensitive to shape and refractive index, and can be sorted in a pulse height analyzer to give the concentration and size distribution. The laser beam is brought to a second, orthogonal line focus at a wire stop at the collection objective. For a collection objective of numerical aperture at the practical limit of $\sim F/3$, the whole of the central diffractive lobe is collected for particles down to $\sim 3 \mu\text{m}$. For smaller particles, the central lobe will spill over the collection objective and the signal will fall below the value proportional to the particle area.

A crucial aspect of this scheme is to ensure that signals are only accepted from particles which traverse a uniformly illuminated control volume at the lens focus. This is accomplished by a combination of spatial filtering and a side-scatter channel which is used to generate gating signals for the pulse height analyzer (See Fig. 1).

A system of this form has been set up on an optical bench using a 1 mW He-Ne laser. A pair of class III, coated, cylindrical lenses of focal lengths 250 mm and 500 mm are used to create the orthogonal line foci at the measurement station and at the line stop. The collection lenses in each arm consist of a pair of 250 mm $F/2.8$ coated, air-spaced achromats, which are well corrected for spherical aberration. The line stop is a 1 mm diameter wire stretched across the diameter of the lens mount in the forward scatter arm. Adjustable slits are used for spatial filtering in each arm. All the components are mounted on carriages sliding on optical rails bolted to an optical table to give accurate, adjustable positioning of the various elements.

Initial optical alignment and measurement of the laser beam properties, such as waist widths and positions, showed that it is essential to take account of the theory of Gaussian beams rather than relying on simple geometrical optics formulae. It was found necessary to pre-expand the laser beam to some 5 mm diameter with a

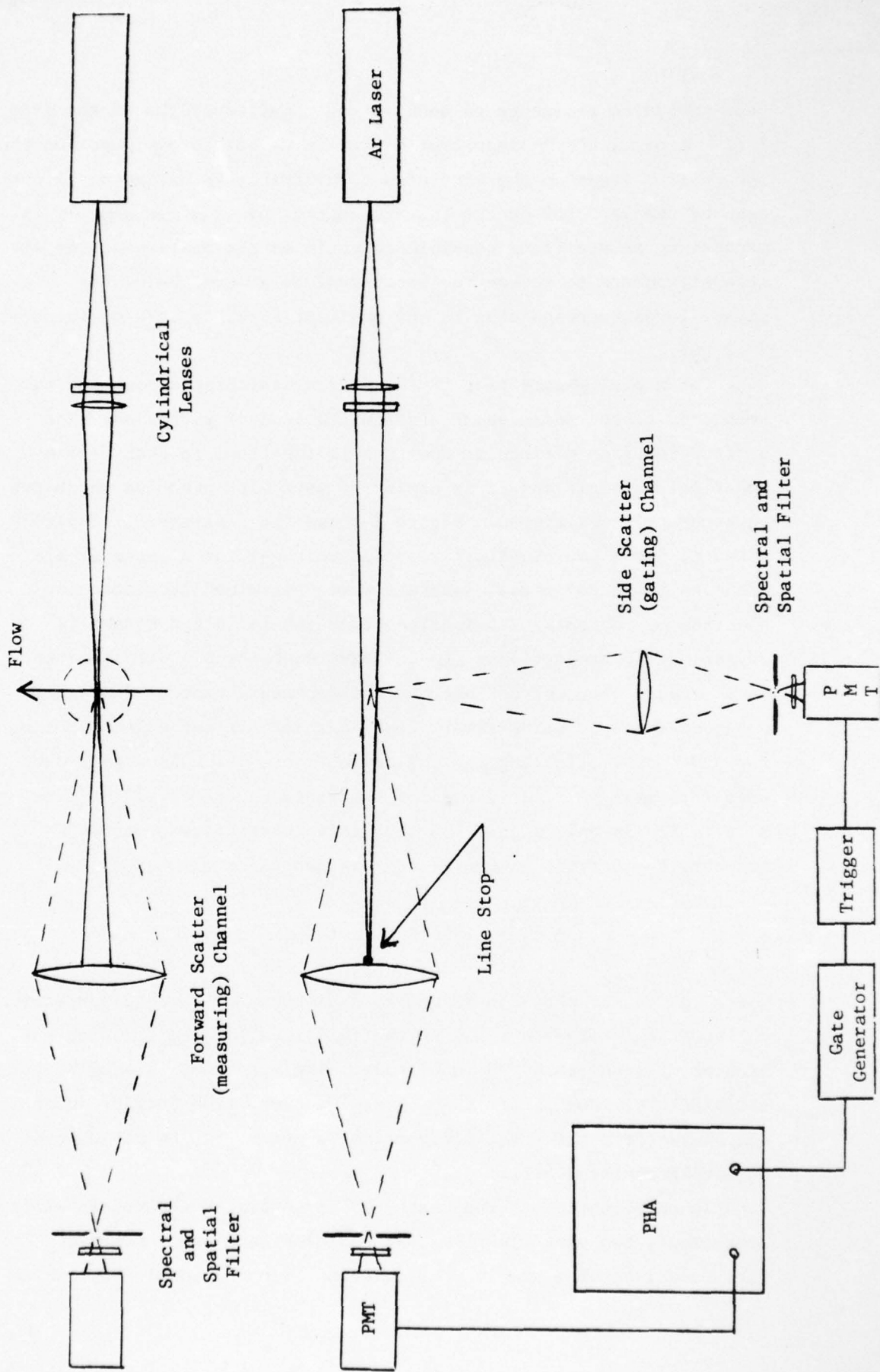


Figure 1. System for Particle Sizing (Schematic)

beam-expanding telescope to achieve the required widths of the line foci. A particularly important matter is to accurately position the second line focus on the wire stop to minimize the background light seen by the detector on the forward scatter arm. After some experimentation, it was found possible to optimize the beam waist and wire stop alignments to reduce the background to a level below the signal level corresponding to the smallest size particle of interest ($\sim 2 \mu\text{m}$).

For a preliminary test of the system, calibrated pinholes have been used at the measurement station, instead of particles. The diffraction from a circular aperture is identical to that from a spherical particle and it is easier to work with pinholes which can be accurately positioned. Figure 2 shows the scattered intensity measured (as a photomultiplier output voltage) for a range of six pinholes (2-50 μm) whose diameters were determined from scanning electron micrographs. It is clear that the collected signal is approximately proportional to the square of the pinhole diameter, as it should be. Corrections for the departure from a square law dependence can be calculated. These account for the effect of the forward lobe overfilling the collection lens, which is significant only for small particles, and for the blocking effect of the line stop, which is only significant for large particles. For our geometry the correction can be written approximately

$$C = 1 - \frac{.64}{d} - .005 d \quad (\text{for } 4 \leq d \leq 50)$$

where d is the particle diameter in microns. With this correction, a theoretical curve is shown as the full line in Fig. 2, which has been normalized on the 25 μm pinhole. The agreement is seen to be satisfactory. For particles $\leq 4 \mu\text{m}$, the correction factor can be calculated from the exact diffraction function, but is not expressible in simple algebraic form.

In anticipation of the next step in testing the particle sizing instrument, two particle flow test vehicles have been prepared. The first consists of a simple recirculating liquid channel flow system

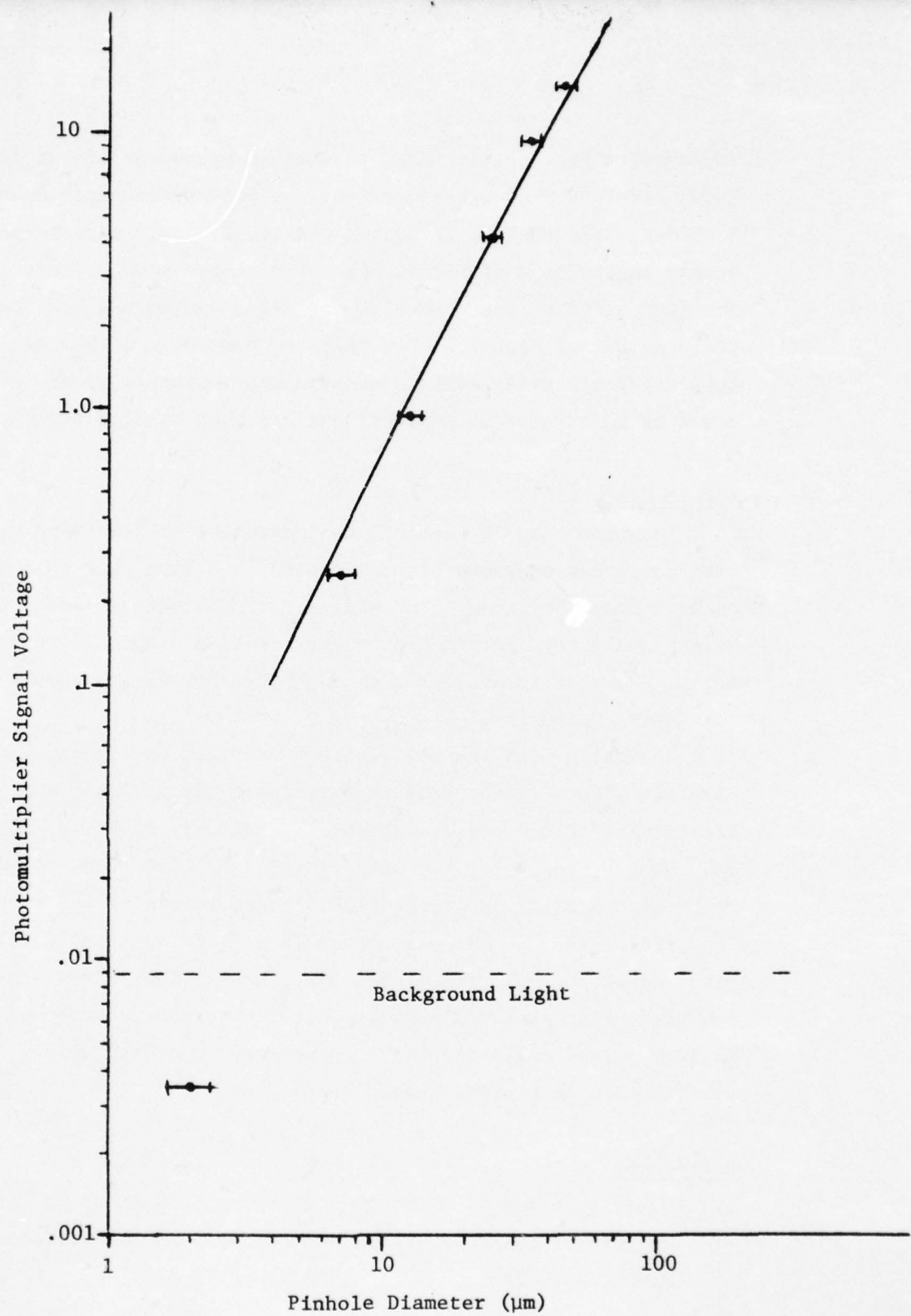


Fig. 2. Photomultiplier Signal Voltage versus Pinhole Diameter.

employing a peristaltic pump, to which dispersions of particles of known size and concentrations, such as polystyrene microspheres, can be added. The channel is square of side 2.5 cm, and has optical access in the form of microscope cover slips cemented over holes in the four sides of the channel. This flow system has been tested and performs satisfactorily. For tests on particles dispersed in air, a commercial vibrating orifice monodisperse particle generator has been acquired under another contract and has been readied for use.

Future Plans

The next step in testing the instrument will be make measurements with particles of known size dispersed in a flow, and to couple the outputs from the forward and side-scatter detectors into a pulse-height analyzer. For initial measurements a simple 128 channel pulse height analyzer is available to us, but a better instrument will be necessary eventually.

Beginning with the new contract year, it is planned to start a parallel effort on the dual focus transit-timing laser anemometer. Preliminary design considerations have already been made in this respect, and involve the use of the strong green and blue lines of an Argon laser with a prism and cylindrical lenses to create two closely-spaced line foci. The two detector channels will be spectrally filtered to give signals deriving from each line focus. The pulse-delay will be measured using standard nuclear instrumentation techniques, involving gating, pulse length to height conversion, and display of the velocity distribution on a pulse height analyzer.

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IV. TURBULENCE

THE COHERENT FLAME MODEL FOR TURBULENT CHEMICAL REACTIONS

TRW Systems
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Dr. J. E. Broadwell, Principal Investigator

As of September 1, 1976, the work on the present contract is approximately 70 percent complete. The status of the work may be summarized as follows:

The coherent flame model has been formulated utilizing the Saffman model for inhomogeneous turbulence as the basis of turbulent diffusivity and turbulent strain rate calculation. The analysis of the turbulent mixing layer, using the model in this form, is complete.

The fluid mechanical calculations for the fuel jet have also been completed and numerical results for the chemical reaction obtained for one case. Based on these results, further study using an integral method is being carried out.

An account of the coherent flame model theory of turbulent combustion is essentially complete. The remaining point has to do with the method of handling the combustion products. A simple example has been formulated to show when dilution of reactants by the products may be important.

All the essential features of the coherent flame model can be derived from first principles with the exception of the flame shortening mechanism which is employed. The correspondence between this process and dissipation in large structures is being investigated.

A no-cost extension of one month has been requested and approval received. With this modification, we expect to complete the research program as planned and within the contracted funds.

LARGE SCALE STRUCTURE AND ENTRAINMENT
IN THE TURBULENT MIXING LAYER

University of Southern California, Los Angeles, California
Subcontract No. 8960-12

Professor F.K. Browand, Principal Investigator
Dr. J. Laufer, Co-principal Investigator
Mr. B.O. Latigo, Research Assistant

Introduction

Previous visual observations^(1,2) indicate the presence of large scale, quasi-organized vortical lumps (vortices aligned across the flow) in the two-dimensional mixing layer. Their existence - documented over a range of Reynolds numbers extending from 10^3 to 10^7 , the highest available result⁽³⁾ - is suggestive of their importance as a characteristic feature of the turbulent flow. As the mixing layer grows downstream, the vortices must necessarily interact to form larger vortices. The interactions to a certain degree are distinct and repeatable. It is precisely these interactions which are responsible for the growth of the mixing layer.

The present experimental study, carried out in a wind tunnel at Reynolds numbers $\sim 10^6$, is intended to provide more information about the large scale structure.

Discussion

Mean Growth Rate

It is important in many practical problems to know how the mean growth rate may be influenced by different initial conditions. It is also possible to learn something about turbulent structure from these measurements. One

of our first tasks has been the determination of momentum thickness at a series of closely spaced downstream stations (maximum Reynolds number $= 1.8 \times 10^6$) for two different initial conditions. One initial condition consists of laminar flow at the trailing edge of the plate dividing the two streams of velocity ratio .23. In the alternate case, a trip wire is placed upstream of the trailing edge on the high speed side - producing a turbulent boundary layer at separation.

The mixing layer with the trip wire grows more slowly in the first 400-500 initial momentum thickness than does the untripped layer. Beyond this point, the tripped mixing layer growth rate approaches the growth rate of the untripped layer - in our case a value of $\theta/x = .0237 \pm .0002$. The untripped mixing layer grows linearly from an origin coincident with the physical origin. There are, however, small, repeatable spatial variations in the downstream growth which are felt to be associated with preferred positions for the occurrence of vortex pairing interactions.

Conditioned Measurements: Detection and Sampling

To isolate the feature of the large scale vortices requires the construction of an ensemble. In our case, the flow is monitored continuously by two detector probes (u-wires) shown in Figure 1, while measurements of u, v are made with our x-wire moving slowly across the flow field. The present data set consists of a continuous record of 10^6 time points (5×10^6 individual digitizations). The mixing layer is tripped, and the measurement station is approximately 1000 momentum thicknesses downstream where the Reynolds number is 1.25×10^6 .

The cross correlation between the two detector probe velocities is

itself an indicator of the underlying coherent, large scale structure. This result is presented in Figure 2. The upper detector velocity (low speed side) is lagged with respect to the lower detector velocity. Minus time indicates early time. Thus, there is a strong negative correlation between the velocities at a slightly early time and a strong positive correlation at a later time. The large scale structure producing this correlation must be skewed - in some mean sense - with the low speed tip leading slightly as shown in the sketch (Figure 2).

In the conditioned sampling technique, the detector signal record is searched for particular occurrences of amplitude and phase. When a particular occurrence is present at the detectors, an origin time is marked, and the flow is sampled for approximately two passage periods (one earlier in time, one later) to produce the initial (or 0th iteration) ensemble. The ensemble is imperfect because the flow is turbulent and, hence, not reproducible in the strictest sense. The detector signals do not unambiguously identify a particular "state" nor are the "states" themselves identical. To sharpen or improve the ensemble, an interactive scheme is employed as shown in Figure 3. Each individual sample is correlated with the ensemble of all samples at a particular y location. The u' , v' , samples are shifted in time to obtain the maximum possible correlation. Samples with low correlation are excluded in the calculation for the new ensemble. The interaction is stopped when the ensemble average reaches a fixed convergence limit. The dramatic improvement achieved by such an interactive scheme is illustrated in Figure 4, for a y position on the low speed side of the mixing layer ($\frac{\bar{u} - \bar{U}}{\Delta U/2} = .85$). Reading from the top, the first two traces are the ensemble averages of the upper (top) and

lower (bottom) detector signals; the third trace is the ensemble averaged perturbation momentum flux; and the last two are, respectively, the vertical and longitudinal velocity fluctuations. The condition was chosen to pick out times when the instantaneous momentum flux is large. The 0th iteration contains 56 samples and the converged iteration contains a subset of 20. Although a much larger sample set must be used to improve the statistical confidence in the ensemble average, the technique seems very promising.

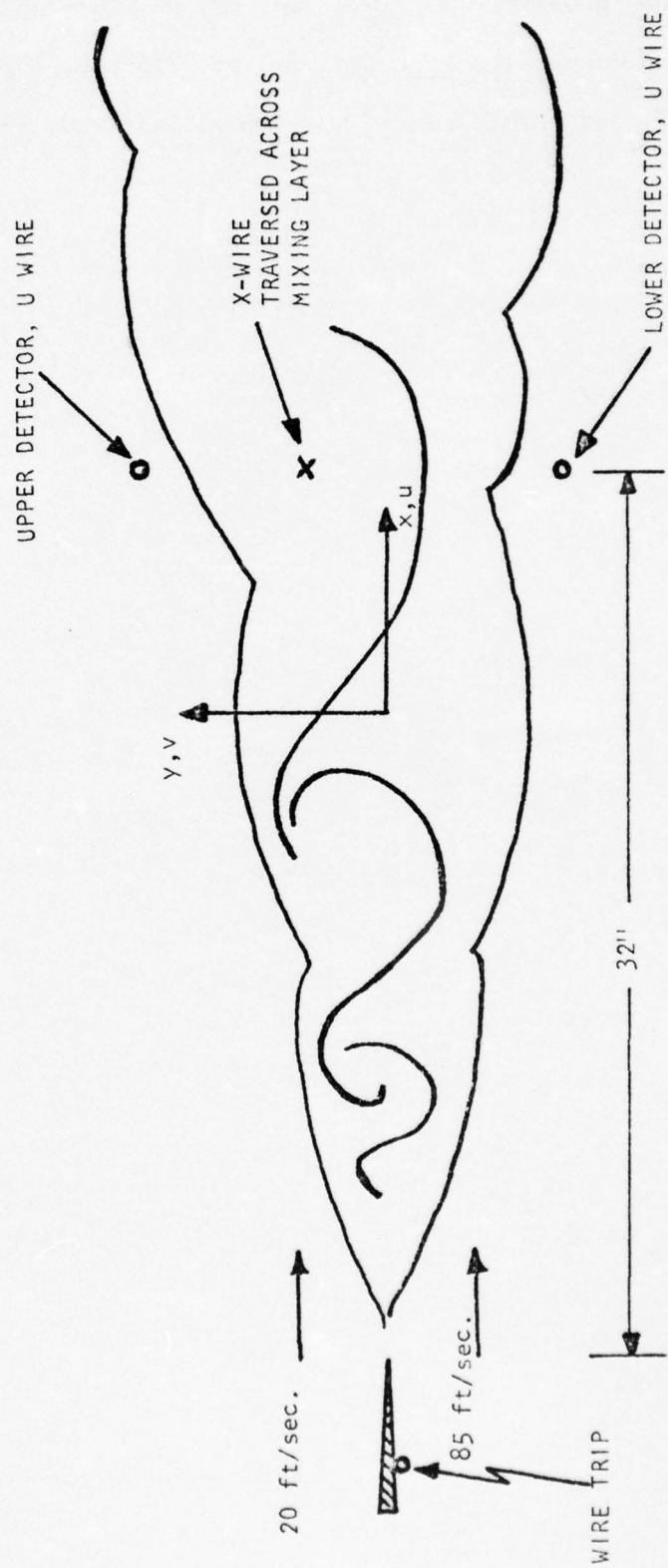
Papers, Presentations

A paper entitled, "Large Scales in the Developing Mixing Layer", by F.K. Browand and P.D. Weidman will appear in JFM, Vol. 76, p. 127, 1976.

The mixing layer growth rate measurements will be presented at the American Physical Society 29th Annual Meeting, University of Oregon, Eugene, Oregon, November 22-24, 1976.

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2. Brown, G.L. and Roshko, A., JFM, Vol. 64, pp. 775-816, 1974.
3. Dimotakis, P.E. and Brown, G.L., SQUID Report LIT-7-PU, 1975.



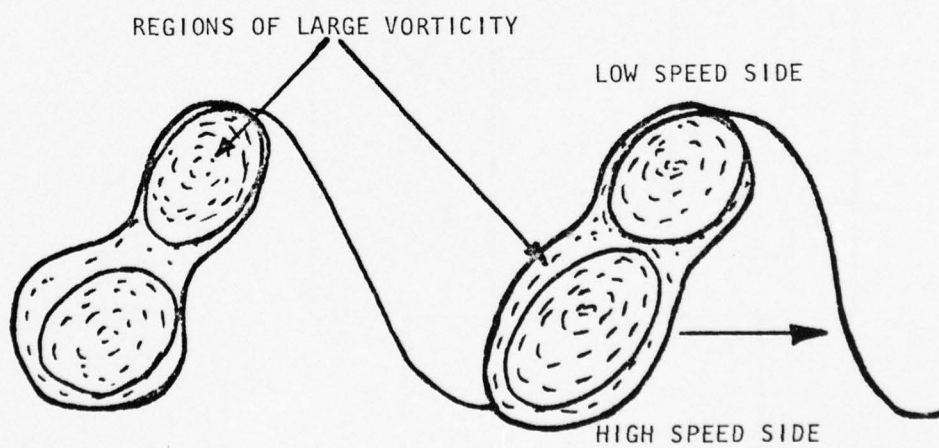
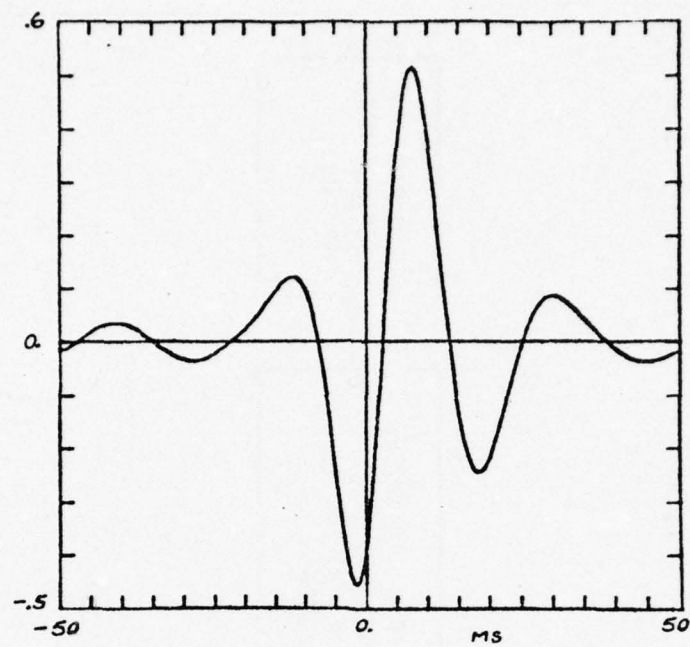


FIGURE 2

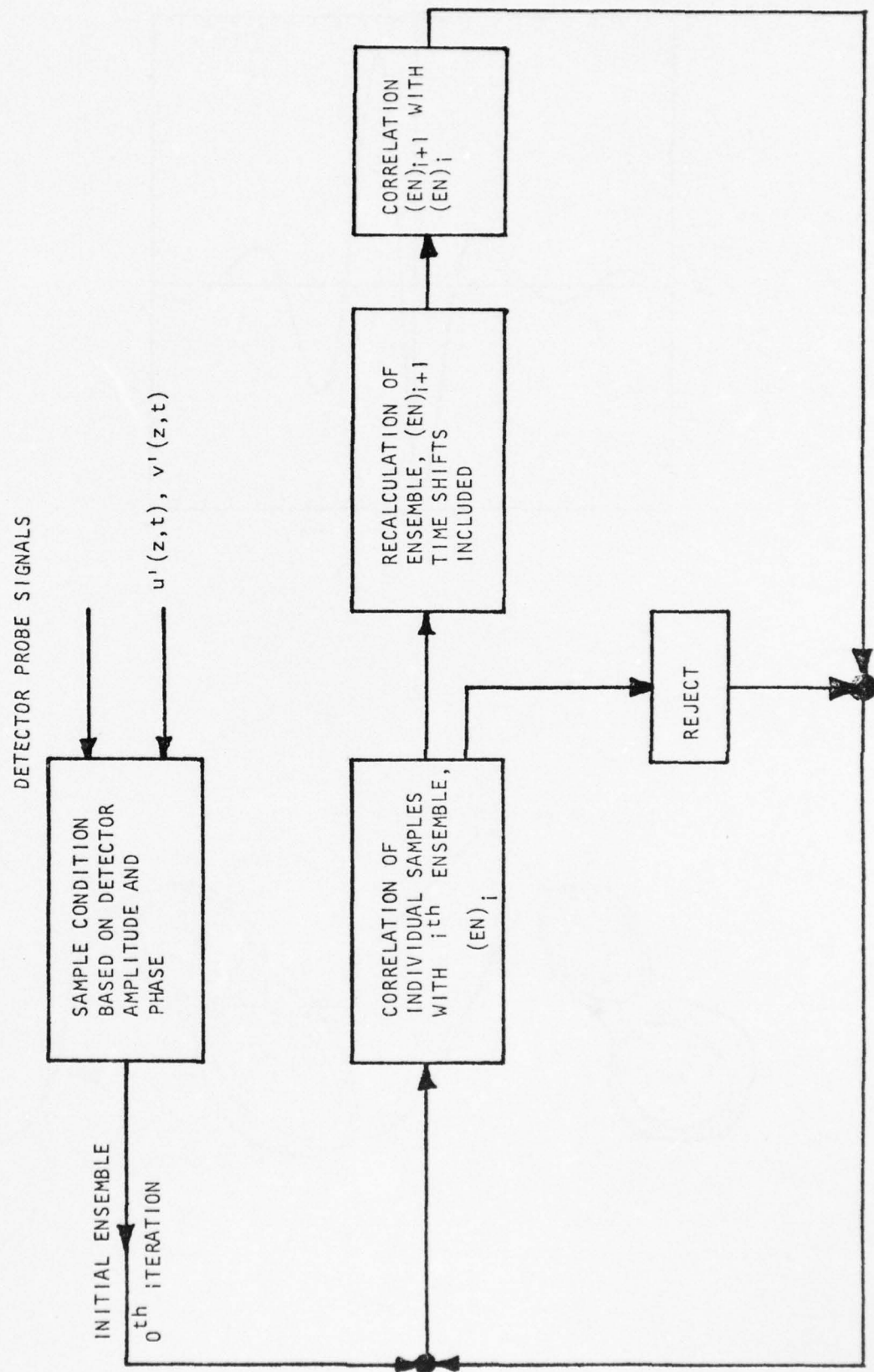
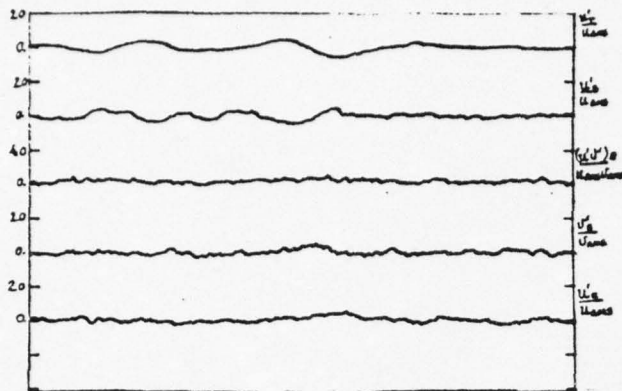
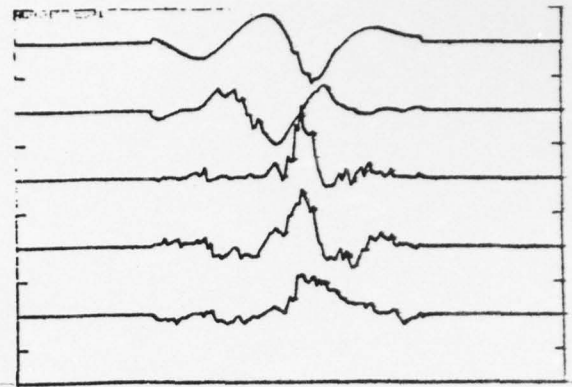


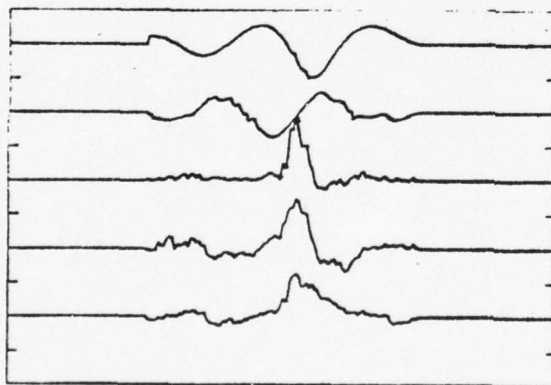
FIGURE 3



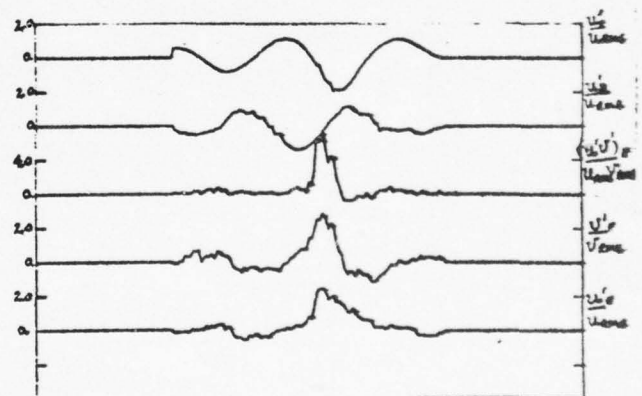
0th ITERATION



1st ITERATION



2nd ITERATION



3rd ITERATION, CONVERGED

FIGURE 4

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BINARY GAS MIXING WITH LARGE DENSITY DIFFERENCE IN HOMOGENEOUS TURBULENCE

Studies of the Basic Phenomena Associated with
Molecular Diffusivity Effects in Turbulent Mixing

Michigan State University, East Lansing, Michigan
Subcontract No. 4964-49

Professor J. F. Foss, Principal Investigator
Mr. K. C. Cornelius, Graduate Research Assistant

Introduction

It is the purpose of our research to illuminate, and to provide quantitative measures of, the fundamental phenomena which are responsible for the strongly enhanced molecular diffusivity effects in a turbulent mixing field. The presence of these effects is of obvious importance in the combustion process; their full exploitation requires an understanding of their dependence upon the character of the turbulence field. One approach toward this understanding is to examine the results of controlled variations in the governing parameters of experiments which are (1) sufficiently simple that the cause/effect relationships are least ambiguous and (2) sufficiently similar to the technological problem that the phenomena of interest are preserved. Our experiments examine the mixing of two distinct rectangular volumes by light scattering measurements from the central region of a closed mixing chamber. The nature of the experimental facility allows the initial turbulence structure in the two volumes to be individually controlled and stable, unstable or neutral density mixing may be investigated.

Each scan of the mixing region can be executed in (\geq) 3.5 msec and each scan can be subdivided into (\leq) 205 segments. The collection optics can be adjusted to examine $7.3 \leq \ell \leq 17$ cm. (Note that for 205 segments at $\ell = 7.3$ cm, the observation volumes are essentially contiguous cylindrical elements with diameters of ≈ 0.25 mm.) The ultimate experimental capability will allow measurements of the non-diffusive mixing of the two volumes, species concentration (Rayleigh scattering) measurements and velocity (L.D.V.) measurements. This binary mixing is considered to faithfully represent the fuel-oxidant mixing pertinent to the combustion process. The homogeneous turbulence condition is congruent with the high Reynolds number condition of technologically important propulsive devices and the consequent "local homogeneity" of the turbulence which is responsible for the molecular mixing therein.

Discussion

The experimental work for the first major study using the mixing chamber and the data collection and storage systems has been completed. The data processing for this study is in progress.

Our specific efforts on this study have been useful in clarifying the appropriate strategies for all studies to be executed with the experimental facility. Namely, the fundamental characteristics of the turbulence field which influence the enhanced molecular diffusivity effects will be inferred from comparative experiments. The experiments to be performed will provide for the direct comparison of two (or more) sets of data for which specific variation (or a limited number of variations) in the control variables has been established. The first study provides a prime example. The specific question which has been addressed is: for homogeneous turbulence fields of

the same energy level and scale, what is the influence of a 6 fold change in the mesh Reynolds number on the non-diffusive mixing if the mesh size and initial velocity are invariant. Implicit in this statement is a constant value for the initial turbulence energy ($q \sim U^2$) and the dissipation rate ($\epsilon \sim q^{3/2}/L$). The two Reynolds number conditions are achieved with the use of Freon 12 and air as the gases; $\nu(\text{air}) = 5.8\nu(\text{Freon 12})$. The Kolmogorov microscale differs by the factor 3.75 for the two conditions during the time period for which the equality of q and L can be assumed. The non-diffusive mixing condition is achieved by contaminating the lower volume with submicron crystals. The crystals are sufficiently large that they only diffuse (approximately) 1/4 of the scattering-volume-span in the lifetime of the experiment and they are sufficiently small that their drag/inertia ratio allows them to faithfully follow the flow.

Preliminary results reveal that the effect on the wave number spectra and average agglomeration lengths are subtle; however, a pronounced effect on the first and second moments of the scalar field spatial distributions are suggested. These matters are under study and will be reported in the Ph.D. thesis of Mr. K. C. Cornelius and the associated report to Project SQUID. It is anticipated that the complementary data from the Rayleigh scattering and L.D.V. measurements for the same experimental conditions will substantially contribute to the understanding which is sought from this research program.

HETEROGENEOUS TURBULENT FLOWS RELATED TO PROPULSIVE DEVICES

University of California, San Diego
Subcontract No. 4965-26

Paul A. Libby
Principal Investigator

Introduction

This research addresses problems related to the turbulent heterogeneous flows which arise in a variety of propulsive devices when reactants and products mix and react. The effort is both experimental and theoretical; the experimental program concerns exploitation and extension of the multiple sensor "hot wire" technique of Way and Libby which permits time-resolved and space-resolved measurements of velocity and concentration of one light species, e.g., helium, in a mixture of light and heavy gases under isothermal conditions. The application of this technique in the present research is to a confined internal flow corresponding to an idealizer combustor. The related theoretical work supports the experimental effort and attempts to extend the results thereof to flow situations of more practical concern, e.g., to chemically reacting flows.

Discussion

During the past six months our theoretical work has involved a continuation of our studies of turbulent reacting flows with particular reference to the case of premixed reactants. Reference 1 which presents

the results of our collaboration with Professor K. N. C. Bray of the University of Southampton and which has been accepted for publication in the Physics of Fluids, concerns the interaction between turbulence and heat release, i.e., not only the frequently studied effect of turbulence on chemical behaviour but also the essentially untreated effect of heat release on the fluid mechanics of the turbulence. The basis for the calculations in reference 1 is the Bray-Moss model for premixed reactants; the specific application is to the oblique, planar reaction zone which has been previously studied in the laboratory by several investigators.

The continuation of this research during the past six months has been in two directions; in one we have attempted to modify the description of the turbulence so as to bring prediction and experimental results in better agreement. It is noted that in one special case the analysis in reference 1 provides an explicit prediction of the angle an oblique reaction zone attains with respect to the on-coming stream of reactants. On the basis of "incompressible" turbulence the angles obtained in reference 1 were considerably higher than those obtained experimentally. However, by including in a heuristic way the effects of variable density, considerably better agreement between theory and experiment has been established. A report on this work is being written.

The second direction in our continuation relates to an extension of the Bray-Moss model to include an appropriate model for probability

density for the velocity components u , v , w . It will be recalled that the original Bray-Moss model related to the pdf of the product concentration c and that the calculation in reference 1 required only that one model pdf. There are two reasons for undertaking an extension to conclude the pdf of the velocity components. The first relates to the possibility of making analytic comparisons of the statistics of all flow quantities within the reaction zone as given by both conventional and Favre averaging. Given the difficulties involved in providing experimental data on such statistics, it is of considerable interest to have analytic estimates therefor even in an idealized flow. At the present time a self-consistent calculation based on the results of reference 1 is being carried out, but we cannot at the moment provide any results.

The second reason for the extension of the Bray-Moss model relates to the possibility of developing an entirely new theory for premixed reacting flows without the gradient assumptions used in reference 1. Gradient or eddy transport assumptions are widely used in predictive methods for turbulent flows, despite the absence of a rational basis therefor. Their use in the thin reaction zone as in reference 1 is perhaps especially suspect; in any case it would be illuminating to have an alternative theory which would not employ the gradient assumption. Such an alternative theory appears possible on the basis of the extension of Bray-Moss, although there are difficulties to be overcome; in particular we are concerned with how the velocity characteristics of unburned parcels of gas within the reaction zone differ from corresponding parcels far upstream of the reaction zone. Similar concerns relate

to a parcel of fully burned gas within the reaction zone and its counterpart far downstream. The analysis of the effects of finite Reynolds number and finite Damkohler on turbulent flame behaviour has been postponed until this alternative theory, which presumably is more fundamental, has been completed.

Our experimental effort during the past six months has been concerned with two aspects of our helium-air hot-wire anemometry. In our previous semi-annual report we presented preliminary results from our experiment related to a two-dimensional helium jet discharging into a moving airstream. These results were promising in the sense that they indicated flow similarity and self-consistency from one of the two probes used in the experiment. During the past few months we have reduced the data from the second probe and have uncovered a discrepancy in the results given by the two probes on the lower half of the jet; we are studying the possible explanation for this discrepancy.

Under partial SQUID support we completed during the past six months and reported in reference 2 an experiment on the slot injection of helium into an turbulent air boundary layer. Of special interest to our SQUID effort is the additional evidence of a counterflux of helium in regions of the boundary layer with significant amounts of helium and thus with significant density fluctuations. It will be recalled that in our previous progress report we noted that such counterfluxes had been observed in our earlier porous tube experiments. A complete explanation of these results is still lacking and is being sought.

During the next six months our experimental effort will be focused on the two-dimensional jet work in order to complete the first phase of this experiment.

References

- 1 Bray, K.N.C. and Libby, P. A., Interaction Effects in Turbulent Premixed Flames. Phys. of Fluids (accepted).
- 2 LaRue, J. C. and Libby, P. A., Measurements in the Turbulent Boundary Layer with Slot Injection of Helium. Phys. of Fluids (submitted)

RESEARCH ON TURBULENT MIXING

California Institute of Technology, Pasadena, California
Subcontract No. 8960-1

Professor A. Roshko, Principal Investigator
Dr. P. E. Dimotakis, Research Fellow
Mr. J. H. Konrad, Research Assistant
Mr. L. P. Bernal, Research Assistant

Introduction

The purpose of this research is to determine the extent of mixing, turbulent and molecular, in plane turbulent mixing layers between streams of different velocities and densities. Central to this effort is the development of techniques for measuring space- and time-resolved concentrations. The research is relevant to problems of chemical mixing, reaction and combustion in turbulent flow. Specifically, we want to define, first, the extent of turbulent mixing for various external conditions such as Reynolds number and, then, the extent of molecular mixing inside the turbulent region. Turbulent mixing sets up the possibility for enhanced contact between fluids from the two sides; molecular interdiffusion then sets up the possibility for chemical reaction. In our experiments the gases are inert so that no chemical reaction actually occurs; the situation may be considered a model for determining the extent of diffusion-controlled, non-energetic reaction between (dilute) reactants being carried by the two streams.

To determine the amount of molecular mixing that has occurred requires, ideally, a concentration sampling probe with perfect resolution (in space and time). From time histories of concentration at various points in the mixing layer, it is possible to determine profiles

of flux of molecularly mixed fluid ("reaction" flux), degree of unmixedness, probability density distributions, and other quantities useful to the engineer or theoretician. An important corollary objective of our research is to develop probes (or other techniques) that can be used to obtain such information.

Discussion

Measurements have now been completed for the following three cases: (i) a mixing layer in which the gases on the two sides are of unequal densities ($\rho_1/\rho_2 = 1/7$) at a velocity ratio of $U_1/U_2 = \sqrt{7}$; (ii) a mixing layer with $\rho_1/\rho_2 = 1$ and $U_1/U_2 = \sqrt{7}$; (iii) a mixing layer with $\rho_1/\rho_2 = 1/7$ and $U_1/U_2 = 1$ (which should more properly be called a wake). In addition to mean velocity and concentration (or density) profiles and corresponding spreading rates, the following quantities have been determined from the time histories of concentration at various points in the profile: r. m. s. concentration fluctuations; intermittency and entrainment; unmixedness factors; probability density functions (pdfs); power spectral densities. The pdfs were used to calculate profiles of reaction product for some idealized, diffusion limited cases of chemical reaction.

An important and unexpected finding is that above a certain critical Reynolds number, the extent of molecular mixing increased by about 20%. From flow pictures it was determined that the transition is due to the appearance of Taylor-like instabilities which themselves become unstable. This cascade introduces into the flow small scale three dimensional structure that is effective in enhancing the small scale and molecular mixing. What is interesting is that the large scale structures and the growth rate of the mixing layer, i. e., the

momentum mixing, are not affected. There are some important implications for modelling and for the problem of turbulent mixing in general. The critical Reynolds number, based on $U_1 - U_2$, the vorticity thickness of the layer, and the kinematic viscosity on the high density (low speed) side, is approximately 2×10^4 .

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2. Konrad, John H. 1976, "An Experimental Investigation of Mixing in Two-Dimensional Turbulent Shear Flows with Applications to Diffusion-Limited Chemical Reactions, " Ph.D. Thesis, California Institute of Technology.

Annual Progress Report, September 15, 1976
SWIRLING HEATED TURBULENT FLOWS AS RELATED TO COMBUSTION CHAMBERS
Project Squid

Mahinder S. Uberoi, Principal Investigator

A theoretical analysis of the structure of a turbulent swirl was made. Velocity and pressure distributions are derived in an infinitely long swirl as functions of space and time by postulating an expression for turbulent stresses and neglecting laminar stresses. Constant angular momentum per unit distance along the swirl axis appears as a parameter.

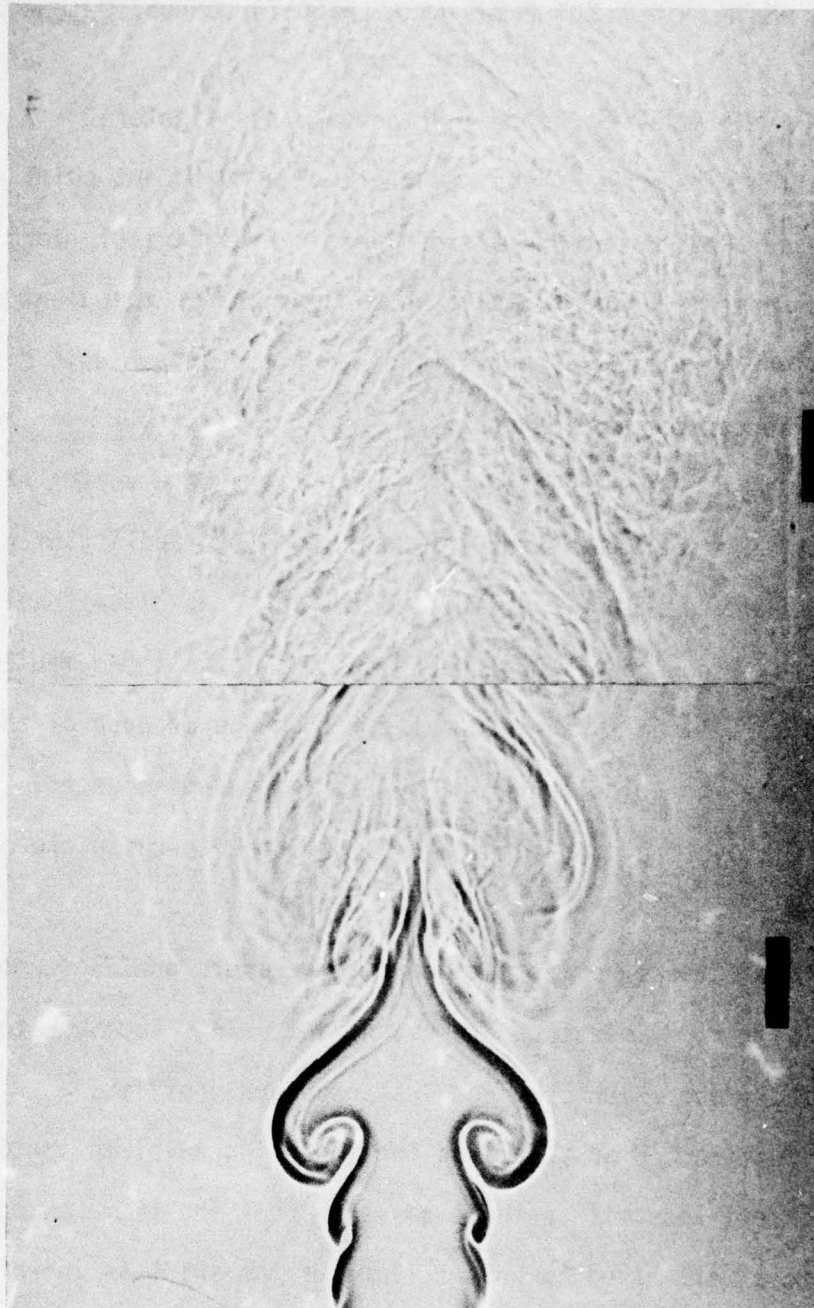
The analysis of experimental work on stability of a vortex was completed. The axial velocity has an important influence on the stability of a vortex.

We have observed initial periodic eddies in a two-dimensional turbulent jet*. Shadowgraphs show a coherent structure at the exit of a two-dimensional jet, which breaks up into turbulence further downstream. The dependence of this structure on the temperature and velocity of the jet is being investigated. The jet temperature is 300° C above the room temperature. We hope to raise the jet temperature to higher values.

We continue to measure instantaneous temperature profiles and some velocity profiles of a warm two-dimensional turbulent jet. This involves the use of our special shooting probe which provides instantaneous profiles.

We have constructed an apparatus for imparting swirl to the 6" diameter gas or air jet. Experiments with natural gas show that the flame changes from a diffusion flame to a well-mixed turbulent flame as the swirl is increased. At high swirls a doughnut-shaped flame sitting close to the gas port is obtained. At still higher swirls the flame is blown out for two reasons: 1) the mixing is so violent that the resulting flow is too lean to burn; 2) at these high speeds of rotation the flame-holding properties of the port are very poor. We are currently designing an electrically heated flame holder.

* See Figure on next page.



Shadowgraph of a 2-D Turbulent Jet. (Uberoi)
Re No $\approx 4 \times 10^3$
 $\Delta T \approx 100^\circ\text{C}.$

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APPENDIX A
Technical Report

SQUID Technical Reports Issued Since 1 April 1976

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MICH-15-PU	Unsteady Transonic Flow with Heat Addition, by Martin Sichel.	February 1976
PU-R2-76	Summary of Activities: Contracts and Reports 1946-1976.	April 1976
UTRC-2-PU	An Experimental Study on the Aerodynamic Response of a Subsonic Cascade Oscillating Near Stall, by F.O. Carta and A. O. St. Hilaire.	August 1976
UTRC-3-PU	Investigation to Extend the Applicability of Laser Raman Scattering Diagnostic Techniques to Practical Combustion Systems, by Alan C. Eckbreth.	September 1976

REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER N/A	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) Project SQUID. Semi-Annual Progress Report, 1 October 1976 1 Apr - 30 Sep 76,		5. TYPE OF REPORT & PERIOD COVERED
6. AUTHOR(s) T./Adamson, E./Bruce J./Broadwell, F./Carta F./Browand, A./Dean		6. PERFORMING ORG. REPORT NUMBER
A. Eckbreth J. Foss J. Fenn M. Lapp A. Fontijn S. Lederman		8. CONTRACT OR GRANT NUMBER(s)
9. PERFORMING ORGANIZATION NAME AND ADDRESS P. Libby J. Ross R. Simpson W. O'Brien W. Sedah J. T'ien A. Roshko S. Self M. Uberoi		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 116p.
11. CONTROLLING OFFICE NAME AND ADDRESS Office of Naval Research, Power Program, Code 473 Department of the Navy Arlington, Virginia 22217		12. REPORT DATE 1 Oct 1976
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19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Semi-Annual Aerodynamics and Turbomachinery Combustion and Chemical Kinetics Measurements Turbulence 4024004		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Reports of progress during the past six months on the 21 research programs comprising Project SQUID are presented. The research programs fall into the areas of Aerodynamics and Turbomachinery, Combustion and Chemical Kinetics, Measurements and Turbulence. Project SQUID is a cooperative program of basic research related to jet propulsion. It is administered by Purdue University and sponsored by the Office of Naval Research.		

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